Disintegration of Selenium-73

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The radiations from the 7.1-hour selenium-73 activity have been investigated using magnetic lens, scintillation pulse-height, and coincidence counting techniques. Two gamma rays of 65.8- and 359-kev energy and of nearly equal intensity are observed to be in prompt coincidence with each other and in delayed coincidence with annihilation radiation resulting from the positron decay of the selenium-73 parent. Energy and intensity measurements indicate that a main group ($E_{max}=1.29$ Mev, $\log ft=5.3$) excites the gamma-ray cascade, while a weaker group ($E_{max}=1.65$ Mev, $\log ft\leq 8.0$) excites the 65.8-kev transition only. These data coupled with internal conversion information lead to a level scheme in As^{73} as follows: ground level, $p_{\frac{3}{2}}$; 65.8-kev level, $f_{5/2}$; and 425-kev level, $g_{9/2}$, $\tau_{\frac{1}{2}}=6$ μsec. The 7.1-hour level in Se^{73} is in a $g_{9/2}$ state.

INTRODUCTION

THE 7.1-hour radioactivity occurring in selenium was first isotopically assigned to Se^{73} by Cowart, Pool, McGowan, and Woodward[1] who bombarded with alpha particles both naturally-occurring germanium and germanium electromagnetically enriched to 90% Ge^{70} and determined relative yields for formation. Scott[2] made a further study of this activity, using a high resolution magnetic spectrometer. He reported a complex positron spectrum and resolved this spectrum into four groups with end-point energies of 0.250, 0.750, 1.318, and 1.68 Mev, with relative intensities 1.1, 10.3, 87.4, and 1.2%, respectively. Four gamma rays were reported with energies of 67.1, 361, 860, and 1310 kev. From internal conversion measurements, Scott suggested that the 67.1-kev transition was of an $M3$ multiple order in the parent Se^{73} and that the 361-kev transition was an $M2$ type following the 1.318-Mev positron transition. The other two gamma rays were presumed to follow the two low-energy positron groups.

Recently, Hooge and Aten[3] reported a 44-minute positron activity assigned to Se^{73} on the basis of threshold and cross section measurements for alpha bombardment of germanium. An end-point energy of approximately 1.7 Mev was determined by absorption techniques and no gamma radiation was observed. The cross section for the formation of the 44-minute activity was about one-fifth of that for the 7.1-hour activity as the alpha-particle energy was varied from 33 to 52 Mev.

The independent-particle model[4] predicts that for nuclei with 39 odd protons or neutrons the two lowest-lying states will be $p_{\frac{1}{2}}$ or $g_{9/2}$ levels with nearly the same energy. Isomeric transitions of the $M4$ type between these two states would be expected. Of the twenty-one known species having 39 odd nucleons of one kind, only four cases are known to have isomeric transitions, all being of the $M4$ type.

[1] Cowart, Pool, McCowan, and Woodward, Phys. Rev. 73, 1454 (1948).
[2] F. R. Scott, Phys. Rev. 84, 659 (1951).
[3] F. N. Hooge and A. H. W. Aten, Physica 19, 1047 (1953).
[4] See, e.g., P. F. A. Klinkenberg, Revs. Modern Phys. 24, 63 (1952).

Se^{73}, containing 39 odd neutrons, would be unique if the $M3$ transition really occurred. The present work was undertaken to investigate the nature of this transition in the light of the known systematics in this region of Z and N.

BETA- AND GAMMA-RAY SPECTRA

The samples of Se^{73} were prepared by irradiating powdered pure germanium metal with 28-Mev alpha particles from the Carnegie Institution of Washington, Department of Terrestial Magnetism, sixty-inch cyclotron. Arsenic and selenium carriers were added to the target which had been dissolved in aqua regia and the material was boiled to near dryness to remove the excess nitric acid and then taken up in $1N$ HCl. Selenium metal was precipitated by the addition of hydroxylamine hydrochloride and potassium iodide, centrifuged, washed, and then used in this form to prepare beta- and gamma-ray sources. The separation was performed a second time for several of the gamma-ray sources since, after several half-lives of the Se^{73} activity, some radiations characteristic of the 26-hour

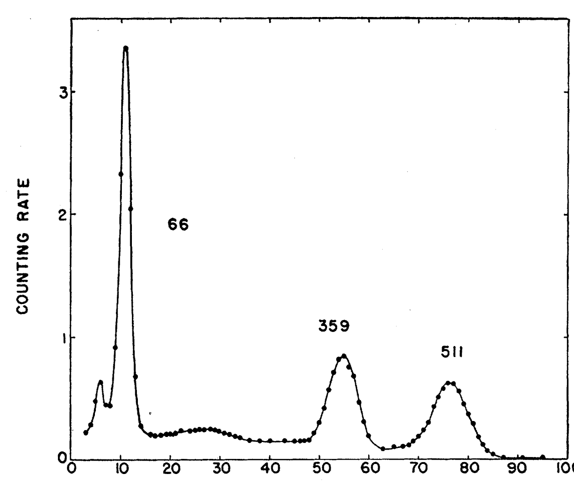

FIG. 1. Scintillation spectrum of the electromagnetic radiation from Se^{73}.

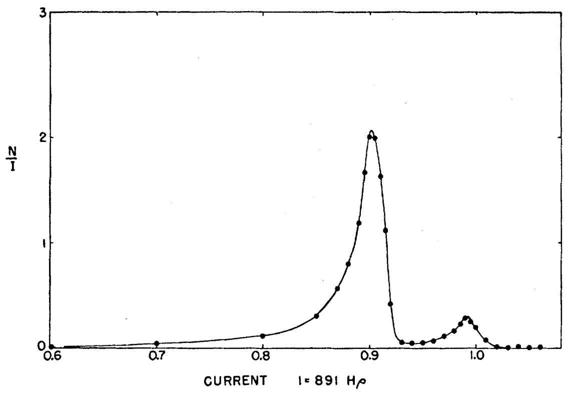

FIG. 2. Internal conversion electrons from the 65.8-kev transition.

As^{72} due to growth from the 9.5-day Se^{72} impurity became a measurable fraction of the total activity.

The electromagnetic radiations emitted by the Se^{73} were examined with a scintillation pulse-height spectrometer employing a thallium-activated sodium iodide crystal two inches in diameter and two inches long viewed by a type 6292 photomultiplier tube coupled to a conventional linear pulse amplifier and a single-channel differential discriminator. The spectrum so obtained is shown in Fig. 1. This spectrum has peaks corresponding to radiations of 66, 359, and 511 kev. The peak at 39 kev is due to the iodine K x-ray escape from the photoelectric capture of the 66-kev gamma ray in the sodium iodide crystal. The relative intensities of the radiations are shown in Table I along with other characteristics to be discussed below. No other gamma rays of an intensity greater than one percent of that for the annihilation radiation were observed. After a period of time the 840-kev gamma ray from the decay of As^{72} could always be found, but would disappear upon further chemical purification of the source.

The conversion electron spectrum and positron spectrum were investigated with a magnetic-lens beta-ray spectrometer having approximately two percent transmission and two precent resolution. The Se^{73} source was mounted on a thin formvar-polystrene film supported by a Lucite ring. The source thickness was appreciable so that some self-absorption and backscattering occurred. The conversion electron peaks occur at 53.96, 64.5, and 347 kev. The first two peaks correspond to the K and L conversion of a 65.8-kev transition in arsenic and the last peak to the K conversion of a 359-kev transition, also in arsenic. The L conversion peak for the 359-kev transition was not well resolved.

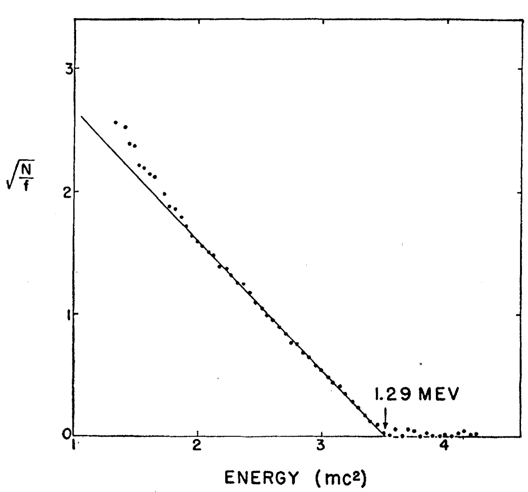

FIG. 3. Fermi plot of the positron spectrum from Se^{73}.

The conversion peaks for the 65.8-kev transition are shown in Fig. 2. The ratio of the number of K electrons to the number of $L+M$ electrons as measured is 10.2 ± 0.2, agreeing closely with other ratios in empirically classified $M1$ transitions and with ratios of calculated K and L conversion coefficients of Rose *et al.*[5] The K to $L+M$ conversion ratio for the 359-kev transition was measured as 8 ± 2, the value being not very accurate since the K and L peaks were not entirely resolved.

TABLE I. Radiations from Se^{73}.

Transition	Energy	Intensity	Half-life
(a) Beta spectrometer measurements			
Positron 1	1.29 ± 0.01 Mev	100	7.1 hr
Positron 2	1.65 ± 0.02 Mev	1	
Int. conv. K_1	53.96 ± 0.1 kev	27.5	
Int. conv. L_1	64.50 ± 0.1 kev	2.7	
Int. conv. K_2	347 ± 1 kev	1.7 } 1.87	
Int. conv. L_2		0.2 }	
(b) Scintillation spectrometer measurements			
Gamma 1	66 kev	82	$<5\times10^{-9}$ sec
Gamma 2	359 kev	100	6.0 ± 0.2 μsec
Annihilation	511 kev	130	

The Fermi analysis of the positron spectrum is shown in Fig. 3. It consists of a main group having an end-point energy of 1.29 ± 0.01 Mev. A weaker group of about one percent of the other group is consistent with the present data, the end point being about 1.65 Mev. Scott, who had a much more intense source available, obtained a better separation from the background and arrived at a similar figure for the intensity. No component was observed having an end point in the region of 0.750 Mev and it is felt that the deviations from the straight-line Fermi plot at the lower energies are attributable to source thickness.

The relative intensities of the conversion electron groups corresponding to the 65.8- and 359-kev transitions and of the positron groups are listed in Table I along with other measured characteristics.

COINCIDENCE MEASUREMENTS

Gamma-gamma and gamma-annihilation radiation coincidences were measured using pulse-height discrimi-

[5] Rose, Goertzel, and Swift (privately circulated tables).

nation in both channels. The pulses from sodium iodide detectors were amplified through pulse amplifiers of 0.1-microsecond rise time and then selected with single-channel differential pulse-height analyzers with a total delay time of less than 0.5 microsecond. These selected pulses were fed to a coincidence circuit of adjustable resolving time, the value used in most of the experiments being one microsecond. The single-channel and coincidence counting rates were recorded as one channel was adjusted to count a particular gamma ray and the entire spectrum was recorded differentially with the other channel. Results of one of the runs is shown in Fig. 4. The coincidence counting rate for the 65.8–359-kev gamma rays has been normalized to the single-channel counting rate for the 359-kev gamma ray. On the assumption that both the 65.8- and 359-kev transitions follow positron decay, one would expect many more 65.8–511 and 359–511 gamma-gamma coincidences than are observed, unless some physical situation were destroying "true" coincidences.

A search was made for delayed coincidences by

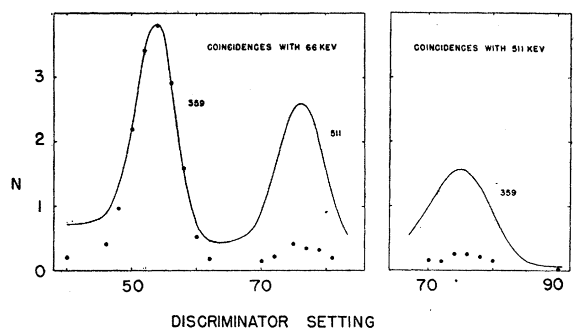

FIG. 4. Gamma-gamma coincidences in selenium-73 as function of energy. Solid line represents single-channel counting rate. Circles represent coincidence counting rate.

introducing additional time delay into one of the channels between the pulse amplifier and the coincidence circuit. Time delays up to about two microseconds were made through the use of RG-65U cable, which has a nominal time delay of 0.042 microsecond per foot. For longer time delays, i.e., up to forty microseconds, an electronic time delay generator was employed. A number of coincidence resolving times up to two microseconds were used. Results of the measurement of delayed positron annihilation radiation with the 65.8- and 359-kev gamma radiation are shown in Fig. 5. The results have been corrected for the random coincidence counting rate and the finite time resolution of the circuit as determined from "prompt" coincidences from annihilation radiation. Both gamma rays are delayed, indicating that they follow the decay of a metastable level having a half-life of 6.0±0.2 μsec. Since the positron spectrum is complex there is a possibility that prompt coincidences may occur between the weaker positron branch and the lower energy gamma ray since the end-point energies of the

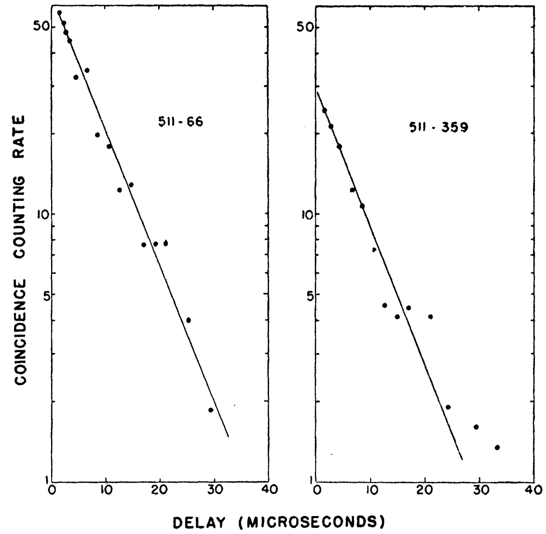

FIG. 5. Delayed coincidences of 65.8- and 359-kev gamma rays with positron annihilation. Resolving time = 1 μsec.

two positron groups differ by about 360 kev. Delayed coincidences were measured between the annihilation radiation and the 65.8-kev gamma ray with a coincidence resolving time of 5×10^{-8} sec, employing delay times from zero to three microseconds. Results of this measurement are shown in Fig. 6. The prompt coincidences represent about one percent of the total integrated delayed coincidences and thus represent an upper limit to the positron branch that can feed the 65.8-kev state. The reason for the upper limit is the possibility of a prompt coincidence between an annihilation radiation pair where one of the pair has been scattered and degraded in energy so as to be recorded as a 65.8-kev gamma ray.

DISCUSSION OF RESULTS

In order to obtain the internal conversion coefficients of the 65.8- and 359-kev transitions, it is necessary to determine the ratio of the number of conversion electrons to the number of gamma rays involved in the particular transition.

It is not possible to make a direct measurement of

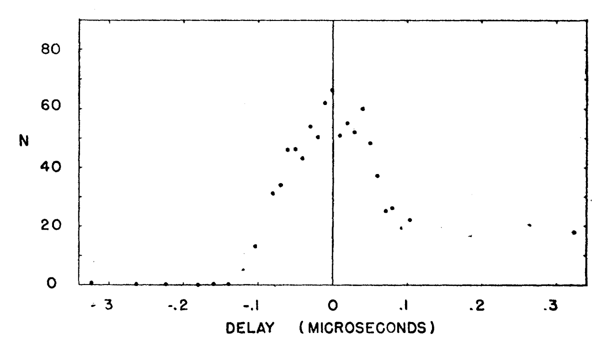

FIG. 6. Delayed coincidences of 65.8-kev gamma ray with positron annihilation radiation. Resolving time = 0.05 μsec.

TABLE II. Half-life and internal conversion coefficients.

Transition	Experimental α_K	Experimental $\tau_{\frac{1}{2}}$ sec		Theoretical α_K	Theoretical $\tau_{\frac{1}{2}}$ sec
65.8 kev	0.22	$<5\times10^{-9}$	$E1$	1.08	10^{-12}
			$E2$	3.00	10^{-5}
			$M1$	0.310	10^{-10}
359	0.011	$(6.0\pm0.2)\times10^{-6}$	$E1$	1.7×10^{-3}	10^{-14}
			$E2$	6.8×10^{-3}	10^{-8}
			$E3$	2.4×10^{-3}	10^{-2}
			$M1$	3.3×10^{-3}	10^{-12}
			$M2$	12×10^{-3}	10^{-6}
			$M3$	46×10^{-3}	10^{-0}

this ratio, but a comparison of the ratio of conversion electrons to positrons as measured with the beta-ray spectrometer with the ratio of gamma quanta to annihilation quanta as measured with the scintillation spectrometer will provide the information required. This method, however, is difficult to apply here, not only because of the wide separation in energy of the quanta involved, but also because of the geometrical uncertainties due to the annihilation of positrons after they have escaped from the source. Another method, experimentally simpler but based on some theoretical assumptions, is to use calculated electron capture to positron ratios to determine the total intensities of the particular transitions. If the rate of population and depopulation of the state are equated, one obtains

$$N_{\beta+}(1+f_{e-}/f_{\beta+})=N_\gamma+N_{e-}.$$

The value of electron capture to positron emission, $f_{e-}/f_{\beta+}$, may be evaluated from the tables of Feenberg and Trigg[6] for the particular positron groups involved. The second method was employed here since the relative intensities of the radiations and coincidence measurements allow a rather unambiguous decay scheme to be determined.

Determinations of $\alpha=N_{e-}/N_\gamma$ based on the decay scheme in Fig. 7 are listed in Table II for each of the transitions. Comparisons with theoretically calculated conversion coefficients[5,7] are made and it is seen that an assignment of a $M1$ multiple order for the 65.8-kev transition is the most likely, the assignment being the same as that derived from the K/L conversion electron ratio. Likewise, the assignment of an $M2$ multipole order for the 359-kev transition is the most likely. The assignment on the basis of the K/L ratio is ambiguous for this energy region and atomic number.

The half-life of 6 μsec for the 360-kev transition also agrees best with the calculated value for $M2$ radiation using the formulas of Blatt and Weisskopf.[8] Values are compared in Table II for the various multipole orders. The effects of internal conversion on the lifetime have been included. An attempt to measure the half-life of the 65.8-kev transition resulted only in an upper limit of 5×10^{-9} sec since the lifetime was too short to be measured by the coincidence circuit.

The ground state of As^{73} is most likely a $p_{\frac{3}{2}}$ single particle state as deduced from the single-particle model, or from the decay of this state to Ge^{73}, or by analogy to the measured spin of As^{75} which differs by two neutrons. The multipole assignments of transitions depopulating the excited states allow the spin assignments to be made as $f_{5/2}$ for the 65.8-kev level and $g_{9/2}$ for the 425-kev level as shown in the decay scheme in Fig. 7.

The value of log $ft=5.3$ for the main positron group leading to the 425-kev state is characteristic of an allowed transition. The $g_{9/2}$ configuration suggested by the shell model is the likely assignment for the 7.1-hour Se^{73} state. The weaker, higher energy positron group having a log $ft\geq8.0$ is compatible with this interpretation since the spin change of two units and a parity change provided by the spin assignment agrees with the observed first forbidden nature of this transition. This disintegration scheme is in disagreement with that reported by Scott.[2]

The 44-minute state decaying with a positron group of 1.7 Mev reported by Hooge and Aten would most likely be a $p_{\frac{1}{2}}$ level. The positron transition to the ground state would be allowed with log $ft=4.8$. Any transition

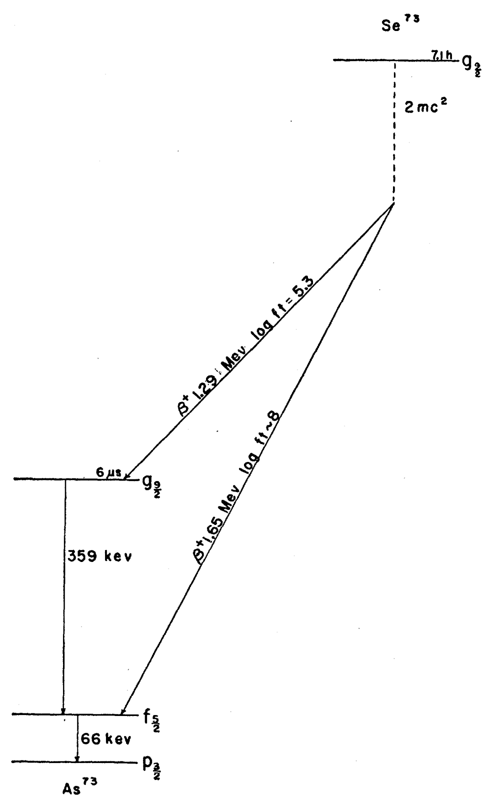

FIG. 7. Disintegration scheme for the Se^{73} 7.1-hour state.

[6] E. Feenberg and G. Trigg, Revs. Modern Phys. 22, 399 (1950).
[7] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report No. 1023, 1951 (unpublished).
[8] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), p. 627.

to either of the higher levels would be twice forbidden or greater and hence could not possibly compete. If the approximate energy measurement of 1.7 Mev is taken at face value, this 44-minute level would lie about 50 kev higher than the 7.1-hour level. The partial lifetime for an $M4$ transition between the two states would be of the order of 10^4 to 10^5 days, based on the Weisskopf lifetime formulas. The branching would be negligible, and thus the decay schemes would be more or less independent of one another.

The authors wish to express their thanks to Dr. N. P. Heydenburg of the Department of Terrestrial Magnetism, Carnegie Institution of Washington, for the use of the cyclotron to make several bombardments and to Mr. A. N. Thorpe for his assistance in making some of the measurements.

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Systematics of Fission Thresholds

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An examination of the experimental data on fission thresholds and ground-state masses of nine nuclei shows that (i) there is experimental evidence for a difference between the masses of even-even, odd-A and odd-odd nuclei in the deformed saddle-point configurations, analogous to the well-known difference for the ground states, (ii) the trend with Z^2/A in the *saddle-point masses* is consistent with simple theoretical estimates based on a liquid drop type of model. This leads to a semiempirical formula for fission thresholds, which is found to be consistent also with estimates of thresholds made with the aid of the observed spontaneous fission half-lives of 28 nuclei. An analogous semiempirical formula for spontaneous fission half-lives is given.

IT is well known that the observed fission thresholds do not show the expected decrease with increasing Z^2/A.[1] The threshold energy is the difference between the mass of the nucleus in its ground state and the mass in the saddle-point configuration[2] through which a nucleus must pass when its excitation energy is only just sufficient to surmount the potential barrier opposing division. It is instructive to examine, separately, the available experimental information on the trends with Z^2/A in these two sets of masses.

Of the lower set of points in Fig. 1 those joined together by lines show the experimental ground-state masses[3] of the nine compound nuclei for which the threshold energies have been measured, as seen from a smooth reference surface $M_{\text{ref}}(A,Z)$, where

$$M_{\text{ref}}(A,Z)-A=-8.3557A+19.120A^{\frac{2}{3}} +0.76278Z^2/A^{\frac{1}{3}}+25.444(N-Z)^2/A +0.420(N-Z) \text{ millimass Units.} \quad (1)$$

This is the semiempirical liquid drop formula for nuclear masses,[4] which follows closely the experimental values except for oscillations associated with shell structure. The nine points exhibit the well-known difference between the masses of even-even, odd-A and odd-odd nuclei (representable,[3] on the average, by a term ± 0.77 mMU in this region of A).

The *experimental* masses of the same nuclei in their saddle-point shapes are shown in the upper part of Fig. 1. They are obtained by adding the threshold γ-ray energy to the ground-state mass in the case of photofission, or the neutron energy and mass to the ground-state mass of the target nucleus in the case of neutron fission. (The neutron binding energy need not be known if the experimental target mass is available.)

It will be noted that there is experimental evidence for a difference between *e-e*, *o-A* and *o-o* masses also in the

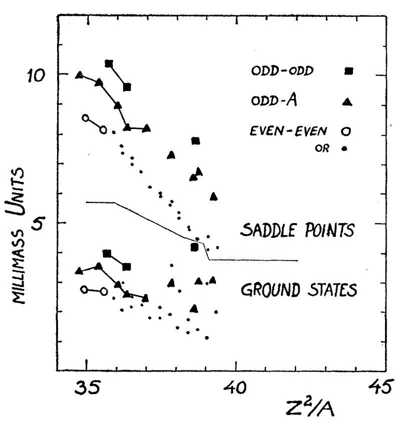

FIG. 1. The masses of nuclei in the ground state and at the saddle point, as seen from a smooth reference surface. All ground-state masses are from reference 3. The nine saddle-point masses shown by points joined together by lines were obtained by adding measured threshold energies (Table I), the remaining ones by adding thresholds estimated from the known spontaneous fission half-lives (reference 8). In the latter case, the threshold of U^{238} was taken as standard for even-even nuclei and Pu^{239} for odd-A nuclei.

[1] See, for example, D. L. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953).
[2] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).
[3] Glass, Thompson, Seaborg, J. Inorg. Nuc. Chem. **1**, 3 (1955).
[4] A. E. S. Green, Phys. Rev. **95**, 1006 (1954).