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PROTON-INDUCED FISSION OF
URANIUM-238 AT MODERATE ENERGY

Larry D. Miller

Leo Yaffe

DOW CHEMICAL U.S.A.
ROCKY FLATS DIVISION
P. O. BOX 888
GOLDEN, COLORADO 80401

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URANIUM-238 AT MODERATE ENERGY**

Larry D. Miller

Research and Ecology

HEALTH PHYSICS RESEARCH GROUP

Leo Yaffe

Radiochemistry Laboratory, Department of Chemistry
McGill University
Montreal, (Quebec) Canada

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PROTON-INDUCED FISSION OF URANIUM-238 AT MODERATE ENERGY

Larry D. Miller and Leo Yaffe

Abstract. Independent and cumulative formation cross sections in the fission of uranium-238 (^{238}U) by protons of energy of 30 to 160 million electron volts (MeV) have been determined for antimony isotopes of mass number 120 to 131. Described are charge-dispersion curves (Z_p = probable charge) constructed and corrected for the variation of the neutron-to-proton ratio of the most probable fission product N/Z_p , as a function of mass (A). The charge-dispersion curves show an asymmetry and are computer-fitted by two overlapping Gaussians. If the curves could be correctly interpreted by the two-mode fission hypothesis, the ratios of asymmetric to symmetric fission at 50 and 160 MeV would be 7.9 and 1.5, respectively. The study is based on a thesis submitted by Larry D. Miller in partial fulfillment of the requirements for the Doctor of Philosophy Degree at the McGill University in Montreal, Canada. Financial assistance was given by the National Research Council of Canada.

INTRODUCTION

Many of the basic features of the distribution of nuclear charge in the thermal-neutron fission of uranium-235 (^{235}U) have been elucidated by the work of Wahl and his co-workers (1).¹ Limited work was done on the problem of charge dispersion at higher energies before the work of Pate, Foster, and Yaffe (2) and that of Friedlander *et al.* (3). These authors studied isotopic distributions and found that, at proton energies below 380 million electron volts (MeV), a symmetric bell-shaped curve was obtained when the yields were plotted against the neutron-to-proton ratio (N/Z) of the fission product. These curves, though broader, are not unlike the isobaric curves, and may be utilized to extract information for comparisons with isobaric data. The most probable charge, Z_p , determined from isotopic experiments, when assigned

to the mass at the peak of the curves (nonintegral) will be identical to the values of Z_p obtained from isobaric curves. Furthermore it is possible to determine a relationship between the full-width at half-maximum (FWHM) of the one curve to the other (4).

A fairly extensive body of data now exists for the proton-induced fission of uranium-238 (^{238}U) in the energy region 20 to 85 MeV. Davies and Yaffe (5) studied the dispersion of cesium isotopes, Parikh *et al.* (6) the dispersion in the mass range $A = 139$ to 143 , and Khan *et al.* (7) investigated yttrium isotopes in the mass range $A = 90$ to 93 . In addition, a number of studies exist in these mass ranges with different fissile targets, such as plutonium-239 (8), uranium-233 (9), uranium-235 (10), thorium-232 (11), and neptunium-237 (12), conducted to determine the role of the neutron-to-proton ratio of the target in determining the charge dispersion.

The following basic trends have been observed:

1. The peaks of the charge-dispersion curves shift towards beta stability with increasing bombarding energy. Mainly, the effect relates to increased neutron boil-off from the target nucleus.
2. The charge-dispersion curves broaden with increasing bombarding energy. This is caused by neutron evaporation from the target nucleus, the fission fragments, and the statistical nature of the neutron evaporation process.
3. A strong correlation appears to exist between the energy at which a certain product will have the greatest probability of being produced, the neutron-to-proton (N/Z) value, and the N/Z value of the fissile target.
4. In the mass range 130 to 140, from the variation of $(Z_A - Z_p)$, where Z_A is the most stable

¹ Numerals in parentheses relate to references at end of text.

charge for mass A , the value of Z_p can be predicted from a knowledge of the value of N/Z of the fissioning target (12, 13). No such evidence exists for the complementary light-mass products ($A = 90$ to 100). The effect has been ascribed to differences in neutron-emitting characteristics of the two fragments.

To date, no systematic correlation has been possible regarding the FWHM values of the charge-dispersion curves at medium bombarding energies. One reason may relate to lack of knowledge of the variation of peak position, N/Z_p , with mass number A . The assumption that the value of N/Z_p is constant with A is not correct as shown by Hogan and Sugarman (4). The assumption is held to be responsible for the fact that the charge dispersion curves at energies around 400-MeV bombarding energy are too broad when compared with those obtained from isobaric studies. Pappas and Hagebø (14) have shown that at 170 MeV, N/Z_p is constant with A only in the narrow region near symmetric fission, $A = 95$ to 115 . However, as will be shown later, plots of N/Z_p against A allow a correction for this variation.

If one corrects for the variation of chain yield with mass, to take into account the slope of the mass-yield curve and the variation of N/Z_p with A , the resulting isotopic dispersion may be compared with the isobaric charge-dispersion curve. This was done by Hogan and Sugarman (4) from the data of Pappas and Hagebø (14) at 170 MeV.

Objectives:

The purpose of the present work is to extend the studies carried out in the laboratory at McGill University concerning the energy-dependence of charge-dispersion curves. The present study in the antimony region was chosen to fill the gap between the yttrium and cesium regions previously studied. This enables the determination of the shape of the function of N/Z_p versus A , assumed to be constant in previous works. The data will also enable a correlation to be found between the widths of charge-dispersion curves and the mass region under investigation. Of interest also will be to determine whether the data obtained would fall on the empirical curve obtained by Friedlander *et al.* (3), as extended by

Davies and Yaffe (5), wherein the energies at which the peaks of the excitation functions of the products obtained were plotted against the neutron-to-proton ratio of the product, resulting in a monotonic function.

EXPERIMENTAL

Targets and Irradiations:

The targets, consisting of a stack of foils described below and mounted on an aluminum target holder, were irradiated in the internal beam of the McGill University synchrocyclotron. The current varied between 1.0 and 1.8 microamperes (μA) and the duration of irradiation varied between 10 to 20 minutes. Two irradiations were performed in the internal beam of the 150-inch synchrocyclotron at the Enrico Fermi Institute, University of Chicago, Chicago, Illinois, arranged by N. Sugarman. The energy range in the McGill University cyclotron used was from 30 to 85 MeV and the energy spread was ± 2 MeV. A nominal proton energy of 160 MeV was chosen for irradiations conducted at the University of Chicago.

The target foils of natural uranium varied in thickness from 0.005 to 0.012 centimeters (cm). All were cut to approximately 1 by 2 cm, with the exception of those in two experiments, in which the foils were trimmed to exactly 2.54 by 5.08 cms with a special die. The target foil was protected against loss due to recoil by aluminum catcher foils (0.0025 cm) placed on either side.

The reactions used to monitor the proton beam were $^{65}\text{Cu} (p, pn)$ ^{64}Cu at proton energies up to 85 MeV and $^{27}\text{Al} (p, 3pn)$ ^{24}Na at 160 MeV. The monitors, high purity copper or aluminum, consisted of a stack of three identical foils (to compensate for recoil losses) placed upstream from the target with respect to the proton beam. The center foil was used to determine the number of protons passing through the target.

Chemical Separation:

Immediately following the end of irradiation approximately 5 millimeters (mm) of the leading edge of

the target stack was sheared off for chemical processing and radiochemical assay. The uranium target foil, together with the aluminum catcher foils, was dissolved in a minimum volume of concentrated hydrochloric acid (HCl), to which a minute amount of hydrofluoric acid (HF) and antimony carrier had been added. An aliquot of this solution was taken for antimony determination.

ANTIMONY (Sb)—The procedure used was a slight modification of the stibine-generation technique described by Greendale and Love (15). This is a rapid one-step procedure based on the Marsh test (16), but with improved chemical yield. The stibine was passed through a tube containing 8-mesh Drierite [to remove contaminants, especially antimony iodide (SbI_3)], decomposed by heat, and collected partly as an antimony mirror on a quartz tube and the remainder as antimony metal on a sintered glass filter. The antimony was dissolved in 6N HCl saturated with bromine (Br_2), boiled to remove excess Br_2 and reduced to Sb^{+3} with sodium hydrogensulfite (NaHSO_3). The solution was boiled to remove sulfur dioxide (SO_2) and antimony trisulfide (Sb_2S_3) was precipitated. The precipitate was filtered, washed, dried, and weighed to determine the chemical yield (usually of order of 70 percent).

When necessary to follow the decay of short-lived isotopes of antimony, or to isolate antimony quickly from its tin precursors, the relatively slow steps involving the antimony mirror and the sulfide precipitation were omitted. The stibine was passed directly into an aqueous solution of silver nitrate (AgNO_3). The resulting mixture of silver (Ag), antimony, antimony oxide (Sb_2O_3), and the compound, Ag_3Sb , was filtered, mounted, and the activity measured. To determine the chemical yields, the activity of shielded ^{126}Sb in these samples was compared to that in a second sample whose chemical yield was ascertained by the sulfide precipitation method described.

COPPER (Cu)—The procedure was based on the elution technique of Kraus and Moore (17). The center copper foil was weighed, dissolved in 2-milliliter (ml) concentrated HCl plus 4 drops 30

percent hydrogen peroxide (H_2O_2), then the solution was evaporated to dryness, the residue taken up in 2-ml 4.5N HCl and absorbed on an anion-exchange resin column (pre-treated with 4.5N HCl). Copper was eluted with 3-column volumes 1.5N HCl, the middle fraction being taken for copper determination. The eluant was diluted to 0.5N HCl, the copper reduced to cuprous ion by boiling with NaHSO_3 and copper thiocyanate (CuSCN) was precipitated with ammonium thiocyanate (NH_4SCN). The precipitate was filtered, washed, weighed, and mounted for activity measurements.

Activity Measurements:

The gamma-ray (γ) spectra were measured with a germanium-lithium [$\text{Ge}(\text{Li})$] detector of 2.0-cubic centimeter (cm^3) active volume coupled to a 1600-channel analyzer. The detector resolution was 4.5 kiloelectron volt (keV) full-width at half-maximum for the cesium-137 (^{137}Cs) 662-keV γ -ray. The detector resolution was sufficient to allow direct determination of several fission products, (e.g., ^{125}Sb) without prior chemical separation.

Treatment of Data:

The formation cross sections were calculated using the monitor cross sections of Meghir (18) and Cumming (19) for copper and aluminum, respectively. The data used are shown in Table I.

TABLE I. Monitor Reaction Cross Sections.

Proton Energy (million electron volts)	^{65}Cu (p,pn) ^{64}Cu ^a (millibarns) ^c	^{27}Al (p,3pn) ^{24}Na ^b (millibarns)
30	390	—
50	210	—
70	160	—
85	140	—
160	—	9.4

^a Copper-65 (proton, proton-neutron) Copper 64, See Reference 18.

^b Aluminum-27 (p,3pn) Sodium-24, See Reference 19.

^c For SI (metric) units: 1 millibarn = 1×10^{-28} square meters.

The methods of calculation used are similar to those of Friedlander *et al.* (3) and will not be repeated. The calculation in the case of the independent cross section of a shielded nuclide or the cumulative cross section of a nuclide with short-lived parents is perfectly straightforward. When the parent decay made a substantial contribution to the activity of the daughter before parent-daughter separation, the timed-separation technique was used with the appropriate growth and decay factors.

Decay Schemes and Nuclear Data:

The nuclear data used are given in Table II. Since most of the antimony isotopes measured are odd-odd nuclei, close to a shell edge, considerable isomerism exists. In all cases, only the longer-lived isomers were measured, so that knowledge of the isomer ratio is required to determine the cross section (σ). These ratios have been determined by

Hagebø (20) for proton-bombarding energies between 159 and 570 MeV. Those for ^{124}Sb and ^{126}Sb are constant and can be extrapolated to the energy region of interest. The isomer ratio for ^{120}Sb is not constant and no extrapolation has been made below 160 MeV.

The cross section for the 60.4-day isomer of ^{124}Sb was converted to the total nuclidic yield by assuming that the isomer ratio of $\sigma_g/\sigma_m = 15.8$, (where g = ground state and m = metastable state) determined at higher energies, is unchanged down to 30 MeV bombarding energy. Similarly, if one takes Hagebø's value, the isomer ratio $\sigma_g/\sigma_m = 4.4$ for ^{126}Sb , and the branching of the isomeric transition is 14 percent. The total cross section is 1.19 times the experimentally-determined ground state (9). Aras and Gordon (21) calculated the isomer ratio for ^{126}Sb produced in thermal-neutron fission of ^{235}U and obtained a value of 4.8, in good agreement with that of Hagebø for the ^{238}U (p,f) ^{126}Sb reaction (p = proton, f = fission).

TABLE II. Pertinent Nuclear Data.

Nuclide	Gamma-Ray Energy (Kiloelectron volts)	Gamma-Ray Intensity (percent)	Half-Life	Isomer Ratio ^c	Remarks
Antimony-120 ($^{120}\text{Sb}^m$)	89.8 197.2	81 ^a 88 ^a	5.76 days ^c	$\frac{\sigma_m}{\sigma_g} = 4.8^d$	At 160 million electron volts
Antimony-122 ($^{122}\text{Sb}^g$)	563.9	66.4 ^a	2.68 days ^c	—	—
Antimony-124 (^{124}Sb)	602.7	98 ^a	60.4 days ^a	$\frac{\sigma_g(3-, 5+)}{\sigma_m(8-)} = 15.8$	—
Antimony-125 (^{125}Sb)	176.4 427.9	7.2 ^a 30.4 ^a	2.71 years ^a	—	—
Antimony-126 ($^{126}\text{Sb}^g$)	666.2 695.1	100 ^a 132 ^a	12.4 days ^c	$\frac{\sigma_g}{\sigma_m} = 4.4$	Isomeric Transition = 0.14 ^a
Antimony-127 (^{127}Sb)	684.9	36.8 ^a	93.4 hours ^a	—	—
Antimony-128 ($^{128}\text{Sb}^g$)	746 752	100 ^b 100 ^b	9.01 hours ^c	—	—
Antimony-129 (^{129}Sb)	543.8	14.9 ^a	4.31 hours ^c	—	—
Antimony-131 (^{131}Sb)	—	—	23.03 minutes ^c	—	—
Iodine-131 (^{131}I)	364.5	79 ^a	8.05 days ^a	—	Descendant of antimony-131
Copper-64 (^{64}Cu)	511	38 ^a	12.8 hours ^a	—	Monitor
Sodium-24 (^{24}Na)	1368.5	100 ^a	15.0 hours ^a	—	Monitor

^a See References 20, 32, 33, 34, 35, and 36.

^b Relative values.

^c See Reference 20.

^d Ground state = g, and metastable state = m.

RESULTS

The cross sections are shown in Table III. The small contribution (less than 10 percent) of the short-lived tin (Sn) isomers to the yield of ^{125}Sb and ^{127}Sb has been neglected. The data given in Table III were extended by those of Panontin and Sugarman (22) at 450 MeV and Hagebø (20) at 570 MeV and 18.2 gigaelectron volts (GeV) to form the excitation functions shown in Figure 1 and the charge dispersion curves shown in Figures 2 and 3. Figure 2 is a typical curve at 70-MeV bombarding energy. It has been constructed by least squares fitting of a Gaussian constrained, so that the independent yields of the isobars for mass-127 sum to the experimentally-determined cumulative yield of ^{127}Sb . The intensity of the γ -rays listed in Table II for $^{128}\text{Sb}^g$ was assumed to be 100 percent, thus providing a limit for the yield. This value has been plotted in Figure 2. From the charge-dispersion curves, interpolation is possible of the yields of ^{128}Sb . Thus an *isomer ratio* will be obtained which is correct only relatively at the various energies. This ratio was found to be constant between 30 and 85 MeV.

Figure 3 is a composite giving charge-dispersion curves from 30 to 160 MeV. Previous workers (5, 6, 7) had used hand-drawn curves, assuming no geometric function, with only the constraint that the independent yields of the isobars, read from the curves, must equal to the sum of the cumulative yields of the isobar, experimentally determined, to which the independently-formed isobars must lead. The curves in this work were assumed to be parabolic in nature and the data fitted by a least-squares computer program. The procedure, while less arduous, leads essentially to the same results.

The choice of a parabola was made since the thermal-neutron data are approximated by a Gaussian function (1). The expression for a Gaussian distribution is:

$$P(Z) = \frac{1}{\sqrt{c\pi}} \exp \left[-\frac{(Z - Z_p)^2}{c} \right] \quad (1)$$

In Equation 1,

$P(Z)$ – fractional independent yield
 c – constant for width of curve

TABLE III. Formation Cross Sections of Antimony Isotopes.

Isotope (i or c)*	Neutron-to-Proton Ratio (N/Z)	Proton Energy	Cross Section (millibarns)															
			30		50		70		85		160		450 ⁽²²⁾	570 ⁽²⁰⁾	18,200 ⁽²⁰⁾			
			(million electron volts)															
120m(i)	1.353		—		0.017±0.002		0.14±0.01		0.33±0.04		1.4±0.2		5.44±0.13		8.23±0.40		2.65±0.09	
120m+g(i)	—		—		—		—		—		1.8±0.2		—		9.50±0.40		3.10±1.0	
122(i)	1.392		—		0.95 ±0.10		3.3 ±0.3		5.1 ±0.5		8.6±0.9		12.3 ±0.2		12.3 ±0.7		2.88±0.24	
124g(i)	1.431		2.6±0.3		10 ±1		18 ±2		18 ±2		15 ±2		11.7 ±0.5		13.6 ±0.6		3.66±0.23	
124m+g(i)	—		2.8±0.4		11 ±2		19 ±3		19 ±3		15 ±2		13.0 ±0.5		14.4 ±0.6		3.85±0.25	
125(a)	1.451		11 ±1		16 ±2		21 ±2		21 ±3		13 ±2		—		—		—	
125(c)	—		17 ±3		24 ±4		48 ±7		35 ±5		20 ±3		—		—		—	
126g(i)	1.471		10 ±1		17 ±2		20 ±2		20 ±2		13 ±1		8.06±0.16		8.52±0.23		2.48±0.11	
126m+g(i)	—		12 ±2		21 ±3		24 ±3		23 ±3		15 ±2		9.26±0.1		10.36±0.23		3.00±0.11	
127(b)	1.490		19 ±3		23 ±3		25 ±4		24 ±3		14 ±2		8.4 ±0.8		8.03±0.06		2.94±0.17	
127(c)	—		33 ±3		33 ±3		33 ±3		33 ±3		20 ±2		12.8 ±0.5		11.3 ±1.8		4.38±0.40	
128g(i)	1.510		14 ±4		14 ±3		13 ±3		11 ±3		—		4.56±0.13		5.00±0.14		1.81±0.06	
129(c)	1.529		14 ±3		11 ±3		9 ±3		8 ±1		12 ±3		3.8 ±0.2		3.01±0.18		1.50±0.06	
131(c)	1.569		11 ±3		7 ±2		7 ±2		7 ±2		5 ±1		—		3.9 ±1.2		2.36±1.1	

*i = independent yield, and

c = cumulative yield (g = ground and m = metastable states).

a = independent plus a small contribution from 9.7-minute tin-125m ($^{125}\text{Sn}^m$).

b = independent plus a small contribution from 4.1-minute $^{127}\text{Sn}^m$.

(20) (22) = See references at end of text.

FIGURE 1. Excitation functions of antimony (Sb) isotopes formed in the fission of uranium-238 by protons. Data above 160 million electron volts are those of Panontin and Sugarman (Reference 22) at 450 million electron volts and Hagebø (Reference 20) at 570 million electron volts and 18.2 gigaelectron volts.

Z — charge

Z_p — most probable charge

When the logarithm of Equation 1 is taken, the expression assumes the form of a parabola:

$$\log P(Z) = -\frac{1}{c}(Z - Z_p)^2 + \left(\log \frac{1}{\sqrt{c\pi}} \right) \quad (2)$$

In the present work, the parameters are cross sections and units of N/Z . The use of the former implies a flat-mass yield curve, an approximation at the lower energies, but certainly correct for this mass region at the higher energies. Equation 2 was expanded to the form of a second-order polynomial for fitting by the least-squares routine. Dispersion curves were also constructed from the data of Panontin and Sugarman (22) and Hagebø (20). These are discussed later.

DISCUSSION

Excitation Functions:

The excitation functions, shown in Figure 1, of the independent-formed nuclides (^{120}Sb , ^{122}Sb , ^{124}Sb , ^{126}Sb , and ^{127}Sb) show the behavior expected of fission products formed at these energies. The data of Panontin and Sugarman (22) at 450 MeV and of Hagebø (20) at 570 MeV and 18.2 GeV bombarding energies have been included in order to obtain the energies at which the excitation functions reach their maxima.

The excitation functions of the two most neutron-deficient isotopes studied (^{120}Sb and ^{122}Sb) reach their peaks (interpolated) at about 1 GeV. The excitation functions for the independent formation

FIGURE 2. Typical charge-dispersion curve, obtained at 70 million electron volts bombarding energy, constructed by least-squares fitting of a Gaussian to data points. The yield for antimony-128 β has been added to obtain its isomer ratio.

of ^{124}Sb , ^{125}Sb , ^{126}Sb , and ^{127}Sb show well-defined peaks within the present energy range studied. The functions clearly indicate also the decrease in energy at which the formation of the nuclide is most probable as the value of N/Z increases. The excitation function for ^{128}Sb and those for ^{129}Sb and ^{131}Sb (actually cumulative, but close to independent because of their early position in the decay chain) reach their maxima at energies below those studied here.

Friedlander *et al.* (3) have shown if one plots the energies at which the excitation functions of cesium (Cs) isotopes reached their maxima versus the value of N/Z of the nuclide that these values fall on a smooth curve. The data for rubidium-84 (^{84}Rb)

and ^{86}Rb fall on the curve as well. Subsequently the curve was extended to lower energies by Davies and Yaffe (5). Parikh *et al.* (6) found that data for barium, lanthanum, and cesium isotopes could be fitted on the curve. The finding was not surprising, since the mass ranges covered did not differ greatly from those of the cesium region.

It is of interest then to note that the data for the antimony isotopes, as shown in Figure 4 fit on the curve drawn from the previous data (6), even though the difference in mass is 10 to 20 units. The position of the antimony points shows that the curve reported by Davies and Yaffe (5) may not be linear at low energies. A forthcoming paper (23) will cover this aspect.

Charge Dispersion:

In Table IV are shown the parameters for the charge-distribution curves. As noted, in this energy range and mass region, correction for the variation of N/Z_p as a function of A is small. Thus Table IV contains the uncorrected data. Figure 5 shows the variation of $Z_A - Z_p$ as a function of energy, values of Z_A being those of Coryell (24). The rapid initial drop, followed by a gradual movement towards stability, as found in other systems, is evident here.

The values of the full-widths at half-maximum increase from 1.6 to 2.5 charge units in the proton energy region: $E_p = 30$ to 160 MeV. In order to determine the effect of the slope of the mass-yield curve on the width of the charge-dispersion curve, the cross-section data were converted to fractional yields, by dividing by the mass yields interpolated from the curves of Stevenson *et al.* (25). These cumulative yields need not be correct on an absolute scale, as long as they agree relatively, so that the slope is not in error. The yield curves (25) show the greatest slope in this mass region at a bombarding energy of 30 MeV. The widths of the charge-dispersion curves increased only slightly when the parabola were fitted to the fractional chain yields, showing that the assumption of a flat mass-yield curve in establishing the curves, even in the area of greatest change, scarcely perturbed the data.

FIGURE 3. Charge-dispersion curves at 30 to 160 million electron volts.

FIGURE 4. Energies at which excitation functions (See Figure 1) reach their maxima as a function of the value of the neutron to proton ratio (N/Z) of the fission product.

Hogan and Sugarman (4) have shown that in order to obtain correct values of FWHM for a charge-dispersion curve, obtained from isotopic rather than isobaric data, the values must be corrected for the variation of N/Z_p with A . It has been shown (26) at higher energies that, in the mass region presently under study, the value of N/Z_p depends strongly on mass. The magnitude of this correction was found using data from previous studies (5, 6, 7) covering mass ranges 90 to 93, 130 to 138, and 139 to 143, as shown in Figure 6. Also shown for comparison is the variation of N/Z_p with A for thermal-neutron fission (1) of ^{235}U and the fission of ^{238}U by 160-MeV protons (26). These curves were used to calculate the value of $N/Z - N/Z_p$ for each isotope of antimony and the charge-dispersion parameters were recalculated. The results, shown in Table V indicate that the isotopic curves are 10 to 19 percent wider than the isobaric curves in this mass and energy region.

Previous workers have used Gaussian or roughly symmetrical curves. Examination of the curve shown in Figure 2 indicates that a more flat-topped curve might also be compatible with the data.

TABLE IV. Parameters of Charge-Dispersion Curves.

Proton Energy (million electron volts)	Peak Position			Difference Between Most Stable Charge (Z_A) and Most Probable Charge (Z_p) ($Z_A - Z_p$)	Full-Width at Half-Maximum	
	Neutron-to-Proton Ratio (N/Z_p)	Proton Mass Charge (A_p)	Most Probable Charge (Z_p)		Ratio (N/Z)	Charge Units (Z)
30	1.498	127.5	51.0	2.0	0.081	1.6
50	1.481	126.5	51.0	1.7	0.084	1.7
70	1.475	126	50.9	1.6	0.095	2.0
85	1.473	126	50.9	1.5	0.107	2.2
100 ^a	1.495	137	54.9	1.4	—	—
160	1.463	125.5	50.9	1.4	0.121	2.5
200 ^a	1.485	137	55.1	1.2	—	—
380 ^a	1.465	135.5	55.0	0.85	—	—
450 ^b	1.425	123.5	51.0	0.64	0.142	3.1
570 ^c	1.413	123	51.0	0.49	0.148	3.2

^aSee Reference 3.

^bSee Reference 22.

^cSee Reference 20.

FIGURE 5. Variation of the difference between most stable charge and most probable charge ($Z_A - Z_p$) as a function of bombarding energy.

Attempts were made to produce such a curve by fitting two overlapping Gaussians by a computer program to the experimental points. In all cases, except for 30 MeV, the chi-square (χ^2) test showed significant improvement for the fit of the double curve over that of the single. The resulting curves are clearly asymmetrical, as shown for example in Figure 7 for the data at 85 MeV. The widths of the asymmetrical charge-dispersion curves, obtained in this fashion, were converted to N/Z units, corrected for $N/Z - N/Z_p$, as read from the second abscissa, (see Figure 7 and Table VI). Again a correction of about 20 percent is evident. The curve at 30 MeV was not appreciably changed either by mass-yield corrections (a broadening of about 0.1 Z units, well within experimental error) or by fitting to an asymmetric curve, but was made narrower by the correction of N/Z_p with A . McHugh and Michel (27) found their isobaric charge distribution curves to be Gaussian, constant in width to 39 MeV, the last being similar to the

results of Pate *et al.* (2), and others. Indeed even the charge-dispersion curve in this work at 50 MeV ($E^* = 52$ MeV) shows only slight asymmetry.

The widths of the charge-dispersion curves observed in the proton fission as a function of energy in the fission of uranium are shown in Figure 8. As apparent from the preceding discussion, none of the proposed corrections will explain the differences in the widths observed in the different mass regions at these energies. Figure 9 shows the variation of the FWHM at energies of 30 and 85 MeV. The dashed portion of the curves is an interpolation, the data currently being obtained at the McGill University Laboratory. The dependence of widths of the charge-dispersion curves upon mass in thermal-neutron fission is inconclusive. Wahl *et al.* (1) assume a constant width for all mass chains,

*Excitation energies.

FIGURE 6. Variation of neutron-to-probable charge (N/Z_p) with mass A as a function of bombarding energy.

FIGURE 7. Charge-dispersion curve at 85 million electron volts fitted to two overlapping Gaussians. (Dashed portion of curves is interpolated.)

TABLE V. Full-Width at Half-Maximum Value Corrected for Variation of Most Probable Neutron-to-Proton Ratio with Mass(A), [N/Z_p(A)].

Proton Energy (million electron volts)	Uncorrected Width [charge units (Z)]	Corrected for N/Z _p (A) [charge units (Z)]
30	1.7	1.5
50	1.7	1.5
70	2.0	1.7
85	2.2	2.0
160	2.5	2.1

^aNeutron-to-most probable charge ratio (N/Z_p) variation with mass.

^bAsymmetric charge dispersion.

TABLE VI. Corrected Charge-Dispersion Parameters.

Proton Energy (million electron volts)	Peak Position		Full-Width at Half-Maximum		
	(N/Z _p)	(Z _A - Z _p)	(N/Z)	(Z-Units)	(correction applied)
30	1.498	1.96	0.071	1.5	a
50	1.481	1.68	0.081	1.7	a,b
70	1.475	1.59	0.10	2.1	a,b
85	1.473	1.55	0.10	2.1	a,b
160	1.463	1.37	0.116	2.4	a,b

TABLE VII. Relative Areas of the Right-Hand Gaussian to the Left-Hand Gaussian as a Function of Energy.

Proton Energy (million electron volts)	Ratio (area of heavy-mass Gaussian) (area of light-mass Gaussian)
50	7.9
70	2.4
85	2.0
160	1.5

while Notea (28), and Strom *et al.* (29) observed a small variation in width.

CONCLUSION

The fitting of two Gaussians to the charge-dispersion data tempts one to interpret the results in terms of two discrete processes, e.g., symmetric and asymmetric fission. Galin *et al.* (30) in the fission of ²³⁸U by 155-MeV protons concluded that 30 percent of the fissions of uranium are asymmetric, while 70 percent are symmetric. At mass 125, the results of Galin *et al.* indicate that the ratio asymmetric to symmetric is 0.2. Symmetric fission should contribute more to the light isotopes of antimony than to the heavy isotopes.

However, as noted from Table VII, this is not the case. If the ratios of the heavy to the light-mass Gaussians are taken as indicative of the relative contributions from asymmetric and symmetric fission respectively, then at 30 MeV the relative symmetric contribution is small, but increases in importance, until the two are nearly equal at 160 MeV.

If the data of Amarel *et al.* (31) were considered for cesium isotopes (A = 128 to 144), formed in the fission of ²³⁸U by 150-MeV protons, and were treated in similar manner, using fractional yields because of the inconstant mass yield in this mass region, the implied ratio of symmetric to asymmetric fission would be 1.2 [in good agreement with the value of 1.0 reported by Galin *et al.* (30) for this region].

FIGURE 8. Dependence of full-width at half-maximum of the charge-dispersion curves on bombarding energy.

FIGURE 9. Variation of the full-width at half-maximum of charge-dispersion curves as a function of mass of the fission product at 30 and 85 million electron volts bombarding energy.

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