

TABLE VI. Ratios of relative cross sections for some reactions of ^{158}Gd and ^{160}Gd .

14-MeV neutrons ^a	14-MeV deuterons ^b	≈ 22 -MeV bremsstrahlung
$(n,\alpha)_{158}/(n,p)_{158}$ = 0.5 ± 0.2	$(d,\alpha)_{158}/(d,\alpha)_{160}$ = 1.3 ± 0.3	$(\gamma,n)_{160}/(\gamma,p)_{160}$ = 700 ± 200
$(n,pn)_{158}/(n,p)_{158}$ = 0.85 ± 0.30	$(d,\alpha n)_{160}/(d,\alpha)_{160}$ = 0.25 ± 0.15	
$(n,2n)_{160}/(n,pn)_{160}$ = 4000 ± 2400		
$(n,2n)_{160}/(n,p)_{158}$ = 600 ± 150		

^a The Gd_2O_3 targets were ≈ 300 mg/cm² thick and were covered with 6-mil nickel foil.

^b The Gd_2O_3 targets were ≈ 10 mg/cm² and were covered with 0.5-mil molybdenum foil.

Fission yields which have been determined²⁸ also support these assignments. Relative cross sections for several

²⁸ W. R. Daniels and D. C. Hoffman, *Phys. Rev.* **145**, 911 (1966).

reactions on ^{158}Gd and ^{160}Gd were calculated from the observed activities of the various product nuclides (Table VI). Appropriate corrections for decay, counting efficiencies, and isotopic abundances of the targets were made.

ACKNOWLEDGMENTS

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Ratio of Symmetric to Asymmetric Fission in the Proton Bombardment of ^{226}Ra

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The ratio of the symmetric to the asymmetric mode of fission of ^{226}Ra produced by protons is studied over the energy range 10–100 MeV. Discontinuities are observed superimposed upon the over-all rise of this ratio with energy, and this is explained by the periodic emission of neutrons prior to the act of fission when such a mechanism becomes energetically possible. A rough estimation is made of the relative cross sections of successive fission reactions after neutron evaporation, and the relative excitation function for the fission of ^{226}Ra by protons is presented for the energy range studied.

In general, it is found that increasing the excitation energy of an easily fissile nuclide, which normally exhibits the "double-humped" asymmetric mass-yield curve, will cause the "valley" of the curve to rise and subsequently become a symmetric peak. At the same time, the center of symmetry of this curve shifts to lower mass numbers. One explanation of this behavior which is receiving much support at present is that there are two modes of fission known, respectively, as symmetric and asymmetric.

The asymmetric mode is prevalent at low-excitation energies and is probably due to the small differences in the binding energies of the last nucleon added at a "closed shell edge" of the resulting fission fragment. The symmetric mode is that which might be expected to occur if such shell effects were absent and hence this mode of fission becomes more predominant as the excitation energy is increased and the small binding-energy differences at shell edges become relatively less important.

The decrease in mass number of the center of sym-

metry of the mass-yield curve is due to the increased number of neutrons which are emitted from the fissioning nucleus as the bombarding energy is increased. The neutrons in excess of those produced in low-energy fission could be emitted from the nucleus prior to fission¹⁻³, after fission²⁻⁴ or after nuclear deformation has occurred but before scission has taken place.

As these neutrons are emitted in a time comparable with the lifetime of the excited fission nucleus, i.e., about 10^{-14} seconds,⁵⁻⁷ it is difficult to establish experimentally whether they are emitted prior to fission or after fission. However, if neutron emission precedes

¹ R. H. Goeckermann and J. Perlman, *Phys. Rev.* **76**, 628 (1949).

² V. I. Goldanskii, E. Z. Terumov, and V. S. Pen'kina, *Doklady Akad. Nauk SSSR* **101**, 1027 (1955).

³ G. N. Harding and F. J. M. Farley, *Proc. Roy. Soc. (London)* **89**, 83 (1956).

⁴ L. Marquez, *Nuovo Cimento* **12**, 288 (1954).

⁵ N. Bohr and J. A. Wheeler, *Phys. Rev.* **56**, 426 (1939).

⁶ I. Halpern, *Ann. Rev. Nuc. Sci.* **9**, 245 (1959).

⁷ W. E. Stein, *Dissertation Abstr.* **23**, 2566 (1963).

fission then the fissioning nucleus would be left with less excitation energy, the deficit being equal to the sum of the kinetic and binding energies of the departed neutrons. The ratio of symmetric to asymmetric fission should then show the effect of the decrease in excitation energy of the fissioning nucleus since asymmetric fission is a more probable process at lower energies. If the neutrons are emitted from the fission fragments then no such effect should exist.

Studies of the change in the symmetric to asymmetric fission ratio for ^{232}Th , ^{238}U , and ^{239}Pu bombarded with protons of increasing energy⁸ showed clearly that the excess neutrons emitted at higher energies were pre-fission as dips or discontinuities were imposed upon the steadily rising curves of symmetric to asymmetric yields with energy, and these occurred at the positions expected from known neutron binding and kinetic energies. These dips become less pronounced as Z or A of the compound nucleus increased. Indeed, for ^{241}Am bombarded by protons,⁹ the dips could not be seen at all. For ^{239}Pu three dips were observable, for ^{238}U five or six and for ^{232}Th six or seven were evident.

The obvious next step was to try another isotope of smaller Z or A . Unfortunately the next isotope of any stability is Bi which is not easily fissile and shows no asymmetric character in its mass-yield curve, so a compromise was chosen by using ^{226}Ra . This has several drawbacks, as will be mentioned below, but it does show some asymmetric character in its fission.

For previous experiments the fission products ^{113}Ag and ^{139}Ba were chosen to represent the symmetric and asymmetric fission modes, respectively, rather than integrating to total yield under the whole mass-yield curve. This had the advantage that the chemical and activity determinations of many isotopes did not have to be interrelated and ensured that the reproducibility depended upon only one standard procedure for each fission mode. The cumulative yields of these two isotopes were found to be directly proportional to their respective fission modes over the whole of the energy range studied. In addition, the chemical separations and purifications of silver and barium were straightforward and results obtained from them were found to be reproducible to within 2%. Unfortunately, for reasons given below, ^{139}Ba could not be used as representative of the asymmetric fission mode of ^{226}Ra , and ^{83}Br was chosen instead.

EXPERIMENTAL PROCEDURE

Protons were used to excite the target nuclei to energies ranging from the fission threshold to the neighborhood of 100 MeV. The use of protons was preferred as these particles could be obtained at a high

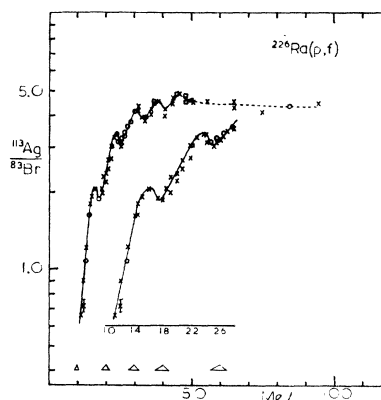


FIG. 1. Ratio of yields $^{113}\text{Ag}/^{83}\text{Br}$ with average proton energy for the fission of ^{226}Ra . \circ Replicate points. \square Points from experiments using the proton linear accelerator.

flux and reasonably monoenergetic over the entire energy range, a requirement that is difficult to meet with neutrons or gamma rays. Deuterons and particles of higher mass were not chosen because of their non-availability over the energy range required for these experiments and because reactions induced by them are more complex due to possible disruption of the bombarding particle.

Irradiations were performed in both the Harwell 110-in. cyclotron and the Proton Linear Accelerator at the Rutherford High Energy Laboratory.

In the cyclotron bombardments the internal probe was used and the thin target was backed by a $\frac{1}{2}$ -in.-thick "Specpure" aluminum block which prevented multiple traversals of the protons¹⁰ and any consequent uncertainty in the energy distribution of the beam. The average energy and spread of the proton beam were determined as described in a previous paper for similar experiments on the fission of ^{232}Th ,¹¹ and the triangles in Fig. (1) show the spread in energy of protons leading to fission for 34% on each side of the indicated energy.

Using the proton linear accelerator, the variation in bombarding energy was accomplished by degrading the spot energies of 30 and 50 MeV by means of aluminum absorbers.

The target consisted of radium chloride painted on to a 0.004-in.-thick "Specpure" aluminum foil to a density of 0.5 mg/cm². This target was wrapped in a double thickness of 0.001-in. "Specpure" aluminum in order to both confine the target and serve as a catcher foil. It was found necessary to use "Specpure" aluminum throughout as the normal grade of aluminum contains a few parts per million of uranium. This contaminant, with its relatively large fission cross section, introduces considerable errors in the determination of fission

⁸ J. P. Butler, B. J. Bowles, and F. Brown, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, New York, 1958), Vol. 15, p. 6.

⁹ B. J. Bowles and F. Brown (unpublished).

¹⁰ J. M. Dickson, Atomic Energy Research Establishment, Harwell Report AERE G/R660 (unpublished).

¹¹ B. J. Bowles, F. Brown, and J. P. Butler, *Phys. Rev.* **107**, 751 (1957).

TABLE I. Comparison of symmetric to asymmetric fission ratios obtained by the catcher-foil technique and total dissolution of ^{238}U target.

Max proton energy (MeV)	$^{113}\text{Ag}/^{139}\text{Ba}$ (catcher foil)	$^{113}\text{Ag}/^{139}\text{Ba}$ (uranium metal)
15	0.208	0.207
40	0.803	0.80
65(1)	1.145	1.15
65(2)	1.167	1.15
100(1)	1.43	1.45
100(2)	1.47	1.45

products due to radium, and these errors become progressively greater as the bombarding energy is decreased.

The double-wrapping technique was used for two reasons, the first being that the target material was rare and had to be conserved for repeated use; the second, that the possibility of contamination of the accelerators by such a hazardous material could not be allowed.

Some doubt was entertained as to the correlation between the results obtained by the catcher-foil technique and those obtained by dissolution of the whole target material, because of the differential ranges of the fission products. Consequently, several bombardments were made at different energies with a target consisting of uranium oxide evaporated on to aluminum and wrapped in the same way as that employed for the radium bombardments, dissolving the wrapping foil only, and the results for these were compared with those from bombardments of uranium metal where the entire target was dissolved. The results, which are given in Table I, show that any differences in the results are less than the experimental error and that it is justifiable to equate the ratios obtained from catcher-foil experiments with those obtained from fission in bulk material.

The contributions to fission from neutrons and gamma rays produced by the total absorption of the protons in the backing material was examined by irradiating a target consisting of two thorium foils separated by aluminum of sufficient thickness to completely absorb the bombarding protons. Two irradiations were performed, one at 10 MeV, the other at 20 MeV. In each case the activity of the thorium on which the beam was incident increased a hundredfold over its natural activity prior to irradiation, whereas the activity of the shielded foil remained statistically the same as an identical unirradiated foil. Hence it was concluded that the contribution of fission by neutrons and photons was negligible in the experiments described here.

The yield of ^{139}Ba , chosen to represent the asymmetric mode in the proton-induced fission of ^{232}Th , ^{238}U , and ^{239}Pu ,⁸ could not be used for ^{226}Ra as the contamination from radium, even using a catcher-foil technique, was sufficient to obscure the barium activity. Attempts were made to remove this contamination from

the very small amount of fission-product barium by using a tandem ion-exchange technique which took about 5 h for a reasonable separation. Since the fission cross section for ^{226}Ra is very small at low energies,^{12,13} activities of only a few counts per minute of ^{139}Ba were expected and these decayed away on the column. The 12.8-day ^{140}Ba could not be used as the activity was too low to count with sufficient accuracy on the counters used and consequently another element had to be chosen as representative of the asymmetric fission mode. The conditions needed to be satisfied were: a half-life short enough to give a fair activity yet not so short that it all decayed away prior to counting, an activity which represented the cumulative yield of the chosen mass chain, rapid and complete exchange with added carriers, a quick and clean chemical separation from other fission products and radium, an easily reproducible form of precipitate and the mass to lie on one of the asymmetric peaks. These conditions proved to be too stringent and a compromise was reached in employing the isotope ^{83}Br to represent the asymmetric mode. This has a half-life of 2.3 h, is easily and quickly separated from other elements and purified and it forms a reproducible precipitate of AgBr . On the other hand, mass 83 lies somewhat on the wing of the light peak^{12,13} and its yield proportional to the total asymmetric yield tends to increase slightly as the bombarding energy, and hence the number of pfission neutrons emitted, is increased.

DEVIATIONS OF THE DETERMINED MASS RATIO FROM THE TRUE MASS RATIO

Examination of the valley of the mass-yield curve for the fission of ^{226}Ra by 11-MeV protons and 22-MeV deuterons¹³ reveals a discrepancy of some 20–30% between the yield of ^{111}Ag and ^{113}Ag , although the general shape of the valley here indicates that these yields should be approximately the same. Similar discrepancies are revealed when the ratio $^{111}\text{Ag}/^{139}\text{Ba}$ from proton-induced fission of ^{232}Th and ^{238}U at 50 MeV and 100 MeV, obtained by Lindner and Osborne,¹⁴ are compared with the ratio $^{113}\text{Ag}/^{139}\text{Ba}$ obtained by Bowles, Brown and Butler,^{10,11} and still further evidence has been obtained by examining the cumulative yields of the mass chains in the vicinity of the center of symmetry of the mass-yield curve resulting from the fission of ^{232}Th , ^{238}U , and ^{239}Pu induced by protons of various energies.¹⁵ This discrepancy, which amounts to some 20% decrease in the yield of ^{113}Ag , is ascribed to the existence of a short-lived isomer as reported by Alexander, Schinde-

¹² A. W. Fairhall, R. C. Jensen, and E. F. Neuzil, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, New York, 1958), Vol. 15, p. 677.

¹³ R. C. Jensen and A. W. Fairhall, *Phys. Rev.* **109**, 942 (1958).

¹⁴ M. Lindner and R. H. Osborne, *Phys. Rev.* **94**, 1323 (1954).

¹⁵ B. J. Bowles and J. P. Butler (unpublished).

wolf, and Coryell.¹⁶ It is apparent that the percentage yield of this isomer is a characteristic of the fission process and does not depend to any great extent on the values of nuclear charge or mass of the target. The correction for this short-lived isomer has not been applied to the results obtained herein as its yield is not known with any degree of accuracy.

Mass 83, as chosen to represent the asymmetric fission mode, does not give a maximum value for the yield of this mode as it lies on the wing of the mass-yield curve. However, neither does mass 139, which has approximately the same yield as mass 83 over the energy range considered here,^{12,13} and there is obviously some difference in the fission of ²²⁶Ra when compared with heavier fissile nuclides. The difference is not so great as to prevent treatment of the data obtained in the same manner as has been done with the nuclides ²³²Th, ²³⁸U, and ²³⁹Pu⁸ provided that one remembers that conclusions drawn from such data are rather less accurate than in those cases. One of the results of taking the yield of ⁸³Br as representing the peak of the asymmetric fission mode of ²²⁶Ra is that the valley-to-peak ratio obtained is high when compared with ratios previously obtained for the more fissile nuclides where ¹³⁹Ba was used to indicate the asymmetric fission yield.

RESULTS AND DISCUSSION

Some 85 determinations of the ratio ⁸³Br/¹¹³Ag were made over the range of 11 to 100 MeV, most of them being between 11 and 50 MeV as this was the range where the discontinuities were expected to be most noticeable. No irradiations were performed at less than 11.0 MeV as the fission cross section is only about 2×10^{-27} cm² at this energy¹³ and decreases very rapidly below this (see Fig. 3). For all but the lowest energy runs, where counting statistics are the overriding criterion, the precision and accuracy of the results is estimated to be about $\pm 5\%$ and $\pm 8\%$, respectively.

The results are presented in Fig. 1. It can be seen, as previously reported for ²³²Th, ²³⁸U, and ²³⁹Pu that, although the ratio of symmetric to asymmetric fission increases with increasing bombarding energy, it does not do so smoothly and that superimposed upon the over-all rise are a number of discontinuities or dips. These discontinuities are most pronounced at low energies and become less prominent as the bombarding energy is increased.

The results are interpreted by the same mechanism as discussed previously, namely, that the ratio of symmetric to asymmetric fission rises as the excitation energy is increased as is well known. However, as soon as the (*p,nf*) reaction becomes energetically possible, the compound nucleus, which is formed on bombardment, can emit a neutron prior to fissioning. This neutron carries away energy equal to the neutron

binding energy plus its kinetic energy and the excitation energy remaining in the resulting nucleus is that much lower than is available for the corresponding (*p,f*) reaction at the same bombarding energy. Consequently the symmetric to asymmetric fission ratio is decreased. As the bombarding energy is increased further, the reactions (*p,f*) and (*p,nf*) occur at higher excitation energies and the symmetric to asymmetric fission ratio rises again until the onset of the (*p,2nf*) reaction produces another dip, and so on. It is evident from the above behavior that neutrons in excess of the so-called "prompt" neutrons, i.e., those produced by increasing the bombarding energy, are emitted prior to the act of fission.

The magnitudes of the discontinuities depend upon the relative probabilities of the onsetting reaction to the sum of the preceding reactions at that energy. The fact that the magnitudes of the discontinuities for the fission of ²²⁶Ra are greater than those for ²³⁸U which in turn are greater than for ²³⁹Pu⁸ and ²⁴¹Am⁹ (for which they are not observed), lends support to the theory¹⁷ that the "fissionability" increases exponentially with Z^2/A of the fissioning nucleus. However, a large proportion of the total reaction cross section for ²²⁶Ra is devoted to spallation reactions^{18,19} and this can readily be usurped by fission reactions, whereas much larger proportions of the total reaction cross sections of ²³⁸U, ²³⁹Pu, and ²⁴¹Am are already devoted to the fission component with correspondingly less room to increase.

The apparent decrease in the ratio of symmetric to asymmetric fission modes as the bombarding energy is increased much above 50 MeV is almost certainly due to the fact that the yield of mass 83 does not represent the peak of the asymmetric mode at low energies. As the bombarding energy is increased, the number of neutrons emitted is increased and, in order to conserve total mass, the light peak of the asymmetric fission-yield curve moves towards smaller masses. This has the effect of increasing the relative yield of the 83 mass chain and thus decreases the ratio of yields of mass 113 to mass 83.

For bombarding energies of 50 MeV and below the ratio of the masses 113 to 83 is represented with negligible error by the ratio of the yields of ¹¹³Ag to ⁸³Br.

It is possible to calculate the energy differences between the onset of each successive reaction giving rise to the discontinuities. The neutron binding energies for the various compound nuclides have been obtained from the compilation of Glass, Thompson, and Seaborg.²⁰

¹⁷ R. E. Batzel, University of California Radiation Laboratory Report No. UCRL-4303, 1959 (unpublished).

¹⁸ R. Vandenbosch and G. T. Seaborg, *Phys. Rev.* **110**, 507 (1958).

¹⁹ R. Vandenbosch and J. R. Huizenga, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, New York, 1958), Vol. 15, p. 688.

²⁰ R. A. Glass, S. G. Thompson, and G. T. Seaborg, *J. Inorgan. Nucl. Chem.* **1**, 3 (1955).

¹⁶ J. M. Alexander, U. Schindewolf, and C. D. Coryell, *Phys. Rev.* **111**, 228 (1958).

TABLE II. Calculated and observed proton energies at the start of the discontinuities. E_n is the binding energy of the last neutron plus 1.1 MeV most probable kinetic energy.

Compound nucleus	E_n (MeV)	Discontinuity	
		Calculated	Observed
^{227}Ac	7.54	8.2	...
^{226}Ac	6.49	14.7	14.7
^{225}Ac	7.83	22.5	22.7
^{224}Ac	6.80	29.3	29.8
^{223}Ac	7.90	37.2	37.2

The average kinetic energy of a neutron evaporated from radium is not available, however it is not expected to be much different than from uranium. This has been measured as being 2.1 MeV from bombardment with 42.5 MeV protons.²¹ From Lang and LeCouteur's equation²² the most probable kinetic energy is half this and a value of 1.1 MeV has been used for the comparison of calculated and observed values as given in Table II. The value for the first discontinuity is not observable experimentally due to the high threshold for the fission of radium but the agreement between the theoretical expectations and experimental results for the other discontinuities leads one to believe that the proposed mechanism is the probable explanation of the observed phenomenon just as for ^{232}Th , ^{238}U and ^{239}Pu .⁸

The gradual disappearance of the discontinuities at higher bombarding energies is partly due to the greater energy spread of the higher energy protons but there are more fundamental causes in addition. Firstly, the rate of change of the symmetric to asymmetric ratio with energy becomes less as the bombarding energy is increased, consequently the difference in the values of the ratio at higher energies is less pronounced than at lower energies. Secondly, the energy carried away by the evaporated neutrons is distributed over a spectrum, and at higher energies there will be an overlapping of reactions such as $(p,2nf)$, $(p,3nf)$, $(p,4nf)$, etc. Thirdly, reactions of the type (p,xnf) are expected to become more improbable at higher values of x , since to undergo such reactions the nucleus must survive against the fission process which can occur at each stage in the neutron "evaporation" process.

RELATIVE PROPORTIONS OF (p,xnf) REACTIONS

Considering these discontinuities, it is observed that they take place over a narrow energy range, 2-3 MeV, after which the curve rapidly resumes its upward trend. This indicates that each reaction $(p,2nf)$, $(p,3nf)$, etc., sets in rapidly and that thereafter the competition does not change greatly until a new reaction becomes possible.

²¹ D. M. Skyrme and W. C. S. Williams, Phil. Mag. 42, 1187 (1951), and private communication.

²² J. M. B. Lang and K. J. LeCouteur, Proc. Phys. Soc. A67, 586 (1954).

The rapid onset of the ensuing reaction can be seen somewhat better by constructing the graph of the logarithm of the symmetric to asymmetric yield ratio versus $(E_{\text{EX}}-5.5)^{-1/2}$, Fig. 2. Here E_{EX} is the excitation energy in MeV and 5.5 MeV is an average value of the fission threshold.²³⁻²⁶ This 5.5 MeV is taken to be the amount by which the nucleus has "cooled" by deforming to a point where fission can occur.

In this case, as with atomic phenomena, the relative probability of two states separated in energy by E may be obtained from the integrated form of the Arrhenius equation, i.e., since the cross sections are directly proportional to the reaction rates,

$$\ln(\sigma_1/\sigma_2) = -E/bT + W, \quad (1)$$

where b is a constant, T is the nuclear temperature, and W is the integration constant.

Bethe²⁷ shows that the nuclear temperature is related to the excitation energy of the nucleus by the equation

$$T = (E_{\text{EX}}/a)^{1/2}, \quad (2)$$

where a is a constant. Hence, referring to the point of scission where the nuclear temperature has cooled to T_f , the equation becomes

$$\log_{10}(\sigma_1/\sigma_2) = -kE/(E_{\text{EXF}})^{1/2} + W', \quad (3)$$

where $k = a^{1/2}/2.303b$ and E_{EXF} is the nuclear excitation at the point of fission, assumed to be $(E_{\text{EX}}-5.5)$ MeV.

The plot of $\log(\text{sym}/\text{asym})$ versus $(E_{\text{EX}}-5.5)^{-1/2}$ should therefore be linear, and it can be seen from Fig. 2 that the graph is linear over the portions where the reaction is predominantly due to one nuclear species.

The linearity of Fig. 2 enables us to observe the onset of the next neutron-emitting reaction more accurately than does Fig. 1 and this graph has been used to derive

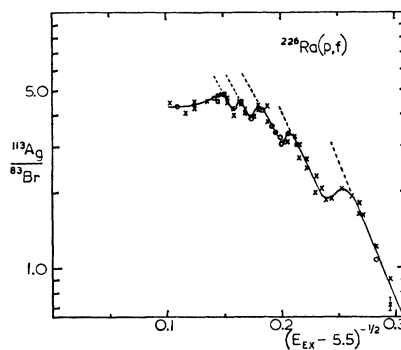


FIG. 2. Ratio of yields $^{113}\text{Ag}/^{83}\text{Br}$ versus $(E_{\text{EX}}-5.5)^{-1/2}$. \circ Replicate points. \square Points from experiments using the proton linear accelerator.

²³ H. A. Tewes and R. A. James, Phys. Rev. 88, 860 (1952).

²⁴ J. L. Fowler, W. H. Jones, and J. H. Paehler, Phys. Rev. 88, 71 (1952).

²⁵ L. Katz, T. M. Kavanagh, A. G. W. Cameron, E. C. Bailey, and J. W. T. Spinks, Phys. Rev. 99, 98 (1955).

²⁶ W. H. Jones, A. Timnick, J. H. Paehler, and T. H. Handley, Phys. Rev. 99, 184 (1955).

²⁷ H. A. Bethe and R. F. Backer, Rev. Mod. Phys. 9, 69 (1937).

estimates of the relative cross sections in the calculations made below.

A roughly quantitative estimation of the relative fission cross sections due to each fission reaction can be arrived at as described in the previous papers.^{8,11} Briefly, at the point where the $(p,2nf)$ reaction has just become established, that is when the curve begins to rise again after the first discontinuity, the symmetric to asymmetric ratio is due to the two reactions (p,nf) and $(p,2nf)$, of which the ratio for the (p,nf) reaction can be estimated by extrapolating the lower linear section of the graph.

If σ_1 and σ_2 are the cross sections for the two reactions (p,nf) and $(p,2nf)$, and the respective fractional yields of the total mass-yield curves are Ag_1 , Br_1 , Ag_2 , and Br_2 , then the observed ratio is given by

$$R = (\sigma_1 Ag_1 + \sigma_2 Ag_2) / (\sigma_1 Br_1 + \sigma_2 Br_2). \quad (4)$$

As the $(p,2nf)$ reaction is occurring at an energy not much above the threshold the yield of Ag_2 is small and, to a first approximation, may be neglected. (Indications from Fig. 1 and the work of Jensen and Fairhall¹³ are that the peak to valley ratio at the commencement of such a reaction is greater than 10 to 1). Further, the Br yields near the peak of the curve are not expected to change much with energy and we can write $Br_1 \approx Br_2$. Equation (4) now reduces to

$$R = \sigma_1 / (\sigma_1 + \sigma_2) (Ag_1 / Br_1). \quad (5)$$

If now r is the value for Ag_1 / Br_1 as obtained from the extrapolated portion of the curve, we obtain

$$\sigma_1 / \sigma_2 = (r - R) / R. \quad (6)$$

The same treatment may be applied at the next discontinuity and so on to give successive values of $\sigma_x / \sum_{n=1}^{x-1} \sigma_n$ and the results of such a treatment are shown in Table III.

The results, though necessarily approximate due to the assumptions made and to the energy spread of the bombarding protons, lend further support to the conclusions drawn in previous publications.^{8,11} The first of these is that the value of $\sigma_x / \sum_{n=1}^{x-1} \sigma_n$ decreases as x increases because of the fact that the fissioning compound nuclei formed at a later stage of the spallation process must have survived against all other fission reactions that are possible during the earlier stages of

TABLE III. Relative cross sections for the (p,xf) reaction, for various values of x in the fission of ^{226}Ra .

x	1	2	3	4	5
$\sigma_x / \sum_{n=1}^{x-1} \sigma_n$	—	1.06	0.38	0.32	0.25

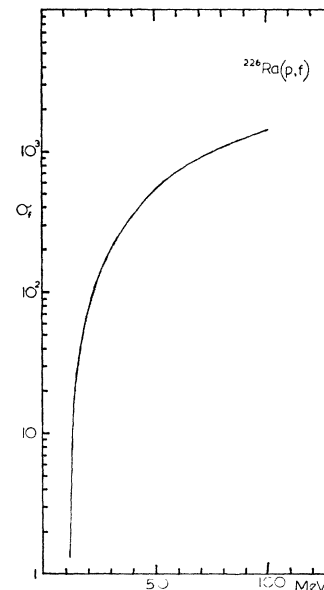


FIG. 3. Excitation function for the fission of ^{226}Ra by protons. σ_f in arbitrary units.

neutron evaporation. The other is that the values from ^{226}Ra are even greater than those obtained from ^{232}Th ⁸ indicating that fission competes with neutron emission on an even less favorable basis than in the case of ^{232}Th .

EXCITATION FUNCTION FOR THE FISSION OF RADIUM BY PROTONS

The excitation function for the fission of ^{226}Ra by protons of up to 100-MeV bombarding energy is given in Fig. 3.

The proton-beam current intercepting the target was monitored by neutron and gamma-ray detectors situated near the probe face of the cyclotron. Thus the relative changes in beam current could be followed accurately although the correction to absolute proton current was known only approximately. The absolute saturation activities of the fission products ^{83}Br and ^{113}Ag were obtained and corrected to the cumulative yields of the masses 83 and 113 where necessary by assuming the unchanged charge distribution rule.²⁸ Using the relative cumulative yields of these mass chains and from published data on the shape of the fission-yield curve at various bombarding energies,^{12,13,29,30} an estimate was made of the area under the whole of the mass-yield curve for each experiment. This gave the relative number of fission events which occurred for a given beam current. The correction for variation in beam current was applied and the results are shown in Fig. 3. The error associated with this curve is estimated to be about 15%, becoming larger at low-bombarding energies.

²⁸ B. J. Bowles (unpublished).

²⁹ R. L. Wolke, Phys. Rev. **120**, 543 (1960).

³⁰ R. C. Jensen and A. W. Fairhall, Phys. Rev. **118**, 771 (1960).