

discussions, Mr. J. Alberghini for his help with some of the measurements and evaluations, and Mrs. M. Speer, Mrs. J. Laraway, and Miss K. McJelton for their help in reducing the large amount of data.

APPENDIX. EVIDENCE FOR THE 1.25-MEV LEVEL

When the 0.6-Mev peak was subtracted from the composite coincident γ spectrum, there was always a remainder (see Fig. 3) which might have been ascribed to the 0.65-Mev γ transition. To check this point more carefully, we measured the γ spectrum simultaneously in coincidence with β particles of 1.2–1.46 Mev, and with β particles of $E_\beta > 1.7$ Mev. That was done by externally routing the coincident pulses into one of the

two halves of the multichannel analyzer, depending on the β pulse height. Since the γ pulses for both spectra went through the same circuits up to and including the analog-to-digital converter, there can be no relative pulse-height shift between the two spectra. Indeed, they matched perfectly at 0.60 Mev. The additional peak was still present after subtraction (see insert of Fig. 3), it appeared with an intensity about 0.7 times that of the 0.72-Mev peak, at an energy of 651 ± 16 kev. In view of the large inaccuracies involved in the subtraction, this is in good agreement with the energy and intensity expected for that transition^{11,13,14} and presents additional evidence for the existence of the 1.25-Mev level in Te¹²⁴.

PHYSICAL REVIEW

VOLUME 121, NUMBER 4

FEBRUARY 15, 1961

Asymmetric Fission of Bismuth*

T. T. SUGIHARA AND J. ROESMER†

Jeppson Laboratory of Chemistry, Clark University, Worcester, Massachusetts

AND

J. W. MEADOWS, JR.‡

Cyclotron Laboratory, Harvard University, Cambridge, Massachusetts

(Received August 24, 1960; revised manuscript received November 7, 1960)

An asymmetric mode of mass division in the mass region 66–73 has been observed in the fission of Bi²⁰⁹ with 36-Mev protons. About 0.3% of the fissions contribute to this mode. At 58 Mev no evidence for asymmetric fission (<0.05% of total fissions) as a separate mode could be found. The fission cross sections at 36 and 58 Mev are 1.9 and 11.3 mb, respectively. The narrowness of the 36-Mev asymmetric peak leads to the suggestion that the asymmetric fission of bismuth results from the fission of a single nuclear species and from a closed-shell effect, similar to the fine structure observed in low-energy fission of heavy elements. This asymmetric fission is considered to occur from states of relatively high excitation energy. However, the possibility of asymmetric fission also occurring from states of low excitation energy, whether following neutron evaporation or as a consequence of an inelastic proton interaction, cannot be ruled out. The symmetric fission observed with both 36- and 58-Mev protons is consistent with the results obtained by Fairhall in the fission of bismuth with 22-Mev deuterons.

I. INTRODUCTION

THE low-energy (<30 Mev excitation) fission of bismuth and radium has been shown by Fairhall and co-workers^{1,2} to be strikingly different in mass distribution from that of thorium and heavier nuclei. Bismuth fission with 22-Mev deuterons¹ results in a narrow, symmetric mass distribution while 11-Mev proton fission of radium² exhibits a “three-humped” distribution. The center peak corresponds closely to the narrow, symmetric bismuth distribution while the

two outside humps resemble the asymmetric modes that might be obtained for thorium fission, suitably adjusted for the difference in mass number. More recently Fairhall, Jensen, and Neuzil³ have shown that symmetric fission is very sensitive to excitation energy but not to target mass number while asymmetric fission exhibits much greater sensitivity to mass number and less to energy.

We felt it conceivable that bismuth might also display an asymmetric mode, no doubt of small probability, if studies could be made at an excitation energy sufficiently low that it is not overwhelmed by the more probable symmetric mode. On the other hand, since the fission cross section drops rapidly with decreasing energy in the particle energy region of 20–40 Mev, there is a practical lower limit as well.

* Supported at Clark University in part by the U. S. Atomic Energy Commission, and at Harvard University by the joint U. S. Atomic Energy Commission—Office of Naval Research program.

† Present address: Nuclear Science and Engineering Corporation, Pittsburgh, Pennsylvania.

‡ Present address: Argonne National Laboratory, Argonne, Illinois.

¹ A. W. Fairhall, *Phys. Rev.* **102**, 1335 (1956).

² R. C. Jensen and A. W. Fairhall, *Phys. Rev.* **109**, 942 (1958); **118**, 771 (1960).

³ A. W. Fairhall, R. C. Jensen, and E. F. Neuzil, *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva*, (United Nations, Geneva, 1958), Vol. 15, p. 452.

In this work initial measurements were made using 58-Mev protons, in which no evidence for asymmetric fission could be found, and subsequently at 36 Mev where definite evidence of a departure from symmetric mass division was observed.

Aside from the work of Fairhall *et al.*,¹⁻³ no low-energy particle-induced mass-distribution studies of bismuth fission had been made at the time these studies were begun, although Sugarman and co-workers⁴ have extensively investigated bismuth fission at energies above 75 Mev. Total cross-section measurements for bismuth fission at energies below 100 Mev have been reported by a number of workers.⁵

II. EXPERIMENTAL

All proton irradiations were carried out at the Harvard University synchrocyclotron whose nominal maximum proton energy is 160 Mev. The proton energies used in this work were obtained by intercepting the internal beam at appropriate radii. The energy was determined by measuring the current on the target probe as a function of the oscillator frequency. Since the phase oscillation amounts to about 1 Mev while the spread in energy due to radial oscillation is about 10 Mev, this method can give a good measurement of the energy and the energy distribution of the beam. The energy values quoted here are most probable values which are about 5 Mev below the maximum value.

Beam currents ranged from 0.1 to 1.0 μ a and bombardment times from 15 min at 58 Mev to two hours at 36 Mev. During the course of this work a total of 22 irradiations were obtained. In most irradiations an internal monitor (Sr^{91}) was used and all formation cross sections were measured relative to it. In two irradiations at each energy the beam was monitored with Cu foil which had been carefully aligned with the target. Absolute measurements of the Cu^{64} activity produced and the known cross sections⁶ for the $\text{Cu}^{65}(p, pn)$ reaction at the two energies (348 mb at 36 Mev, 254 mb at 58 Mev) placed the Sr^{91} cross section on an absolute scale. In these bombardments a 1-mil Al foil was inserted between the copper and bismuth to prevent recoil of spallation products from copper into the bismuth.

Targets were prepared from Bi metal obtained from several different sources. Of these a sample from the Fielding Chemical Company⁷ was found by activation analysis to contain the smallest amount of uranium and zinc impurities, either of which would seriously inter-

fere with the measurements. Details of the impurity analysis are given in Part III. The lumps of Bi metal were fabricated into foils by a method described by Bell and Skarsgard.⁸ Target thicknesses were about 100 mg/cm² and dimensions about 1 cm by 2 cm. The energy loss of a 36- or 58-Mev proton in the target is about one Mev.⁹ The Bi foil was sandwiched between the assembly such that the proton beam was perpendicular to the 2-cm² area and first passed through a guard foil before striking the bismuth. The edges of the guard and target foils were aligned by cutting all three with a razor blade after clamping in the holder.

The guard foils were occasionally counted for recoil loss of Bi fission fragments. This never amounted to more than 0.5%.

After irradiation at Harvard University, the Bi target was chemically processed at Clark University, and separated fractions were counted. A brief description of some of the chemical separation methods is presented in the Appendix. Because many of the cross sections measured were of the order of a microbarn, special pains were taken to insure good decontamination from spallation products formed with cross sections 10⁵ times larger.

Counting measurements were made chiefly on flow beta Geiger counters of a type that has been described.¹⁰ For measurement of very weak activities (less than 50 dis/min) the counters were operated inside 7 in. of steel and anticoincidence shielding. Under these conditions the background rate was 0.25 count/min. In general nuclides were identified by chemical behavior, half-life, and beta absorption curves. In no case were there any appreciable differences from literature data.¹¹ In a few cases with adequate counting rate, radiochemical purity checks were made with a scintillation spectrometer. Observed beta counting rates were converted to disintegration rates by application of correction factors described previously.¹²

III. RESULTS

The formation cross sections measured are listed in Table I and shown graphically in Fig. 1. The values shown have been corrected for chain yield (except for shielded or directly formed nuclides), that is, for the direct formation of isobaric nuclides of higher atomic number than the nuclide counted. Only in the cases of Ni^{66} , Zn^{71} , Zn^{72} , and Ga^{73} was there appreciable (>10%) correction for this effect. The correction was calibrated with the four measured "independent" yields

⁴ L. Jodra and N. Sugarman, *Phys. Rev.* **99**, 1470 (1955); P. Kruger and N. Sugarman, *Phys. Rev.* **99**, 1459 (1955).

⁵ E. Kelly and C. Wiegand, *Phys. Rev.* **73**, 1135 (1946); J. Jungerman, *Phys. Rev.* **79**, 632 (1950); H. Steiner and J. Jungerman, *Phys. Rev.* **101**, 807 (1956); J. T. Gilmore, University of California Radiation Laboratory Report UCR-L-8369, 1958 (unpublished).

⁶ J. W. Meadows, Jr., *Phys. Rev.* **91**, 885 (1953).

⁷ Hudson Heights, New Jersey.

⁸ R. E. Bell and H. M. Skarsgard, *Can. J. Phys.* **34**, 745 (1956).

⁹ W. A. Aron, B. G. Hoffman, and F. C. Williams, U. S. Atomic Energy Commission Report AECU-663, 1951 (unpublished).

¹⁰ T. T. Sugihara, R. L. Wolfgang, and W. F. Libby, *Rev. Sci. Instr.* **24**, 551 (1953).

¹¹ D. Strominger, J. M. Hollander, and G. T. Seaborg, *Revs. Modern Phys.* **30**, 585 (1958).

¹² T. T. Sugihara, P. J. Drevinsky, E. J. Troianello, and J. M. Alexander, *Phys. Rev.* **108**, 1264 (1957).

TABLE I. Bismuth fission yields^a with protons.

Mass number	Nuclide counted ^b	36 Mev (μ b)	58 Mev (μ b)
66	55-hr Ni \rightarrow 5-min Cu ^c	0.48 \pm 0.08	...
67	61-hr Cu	0.72 \pm 0.14	5.5 \pm 0.8
71	3-hr Zn	0.80 \pm 0.04	9.5 \pm 3.8
72	49-hr Zn \rightarrow 14-hr Ga ^c	0.85 \pm 0.05	15.4 \pm 2.9
	14-hr Ga (direct)	0.15 \pm 0.01	2.5
73	5-hr Ga	0.44 \pm 0.03	17
77	39-hr As	0.76 \pm 0.13	35 \pm 5
82	36-hr Br (shielded)	0.26 \pm 0.03	5.9 \pm 0.9
83	2.3-hr Br	5.7 \pm 1.0	123 \pm 8
86	18.6-day Rb (shielded)	2.7	5.1
89	53-day Sr	35 \pm 5	318 \pm 12
91	9.7-hr Sr	56 \pm 3	490 \pm 20
99	67-hr Mo	167 \pm 3	790 \pm 50
111	7.5-day Ag	93 \pm 9	590 \pm 60
112	3.2-hr Ag (from parent) ^d	88 \pm 9	520 \pm 80
	3.2-hr Ag (direct)	8.1	38 \pm 6
113	5.3-hr Ag	85 \pm 13	380 \pm 40
Total fission cross section		1900	11 300

^a Corrected for fractional chain yield unless cross section is for directly formed or shielded nuclide; errors indicate only mean deviation of replicate determinations. All yields measured at least twice unless shown without error.

^b Observed half-lives are given.

^c Counted equilibrium mixture. In some runs Ga⁷² was milked from equilibrium Zn⁷²-Ga⁷² mixture to measure Zn⁷² yield.

^d Ag¹¹² measured in equilibrium with 21-hr Pd parent; yield is that of parent.

(Ga⁷², Br⁸², Rb⁸⁶, Ag¹¹²) and calculated from the equal-chain-length model as described by Pappas.¹³ The smooth curve is drawn above the measured Zn⁷¹ yield since the yield of only one of an isomeric pair was measured.

The errors shown are the mean deviation from the mean of replicate experiments, in general less than 20% and frequently much smaller. Yields measured only once are indicated without an error. Summing the mass distributions of Fig. 1 at 36 and 58 Mev leads to total fission cross sections of 1.9 and 11.3 mb, respectively. On an absolute basis, the error in individual formation cross sections is probably of the order of 30%, not including the error in the monitor cross section. On a relative basis, however, the 36-Mev data should be valid to 15-20% and those at 58 Mev to 10-15%. By this we mean that the ratio of $\sigma(\text{Cu}^{67})/\sigma(\text{As}^{77})$ at 36 Mev should have a root-mean-square error of not more than 30%. The total fission cross sections are probably correct to about 40%, excluding the error in the monitor cross section.

The possibility that the structure in the 36-Mev mass distribution in the neighborhood of mass 66-77 arises from impurities in the Bi target was investigated in some detail. A false peak in this region could result from reactions of two kinds of impurities: fission of elements like uranium and thorium, and spallation of elements like zinc and germanium which are not unlikely contaminants of bismuth.

The uranium-thorium content was measured by irradiating a Bi target about 3 Mev thick (\sim 140

¹³ A. C. Pappas, U. S. Atomic Energy Commission Report AECU-2806, 1953 (unpublished).

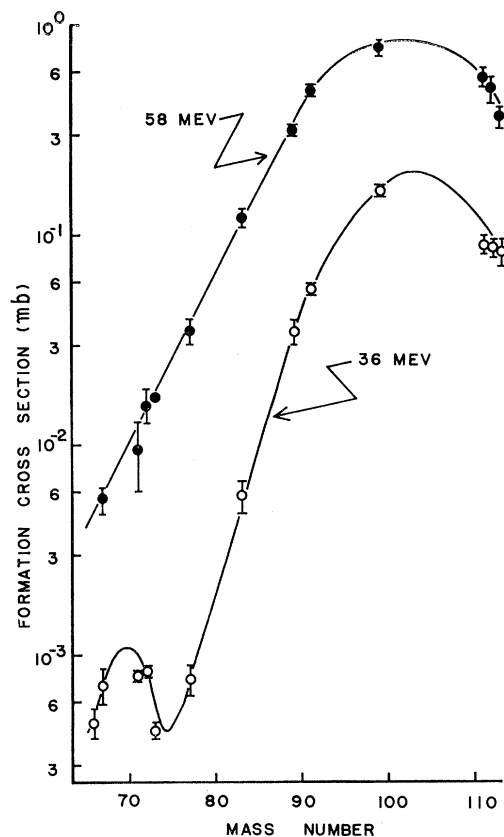


FIG. 1. Mass distribution in fission of Bi²⁰⁹ with 36-Mev (open circles) and 58-Mev (closed circles) protons. Error flags refer to mean deviation of replicate experiments. All yields are corrected for fractional chain yield. The curve is drawn above the point for mass 71 since only one of a pair of isomers was measured.

mg/cm²) with 14-Mev deuterons at the Massachusetts Institute of Technology cyclotron. The beam current was monitored by a thermocouple that had been calibrated with a Faraday cup. Subsequently, strontium and barium fractions were isolated and counted for Sr⁹¹ and Ba¹³⁹. Fairhall has shown¹ that the cross section for Bi fission under these conditions is less than 10⁻³¹ cm², and in any case Ba¹³⁹ is a most unlikely product in Bi fission.¹ Assuming that the fissionable species were all uranium nuclei, we estimated the uranium content from the known formation cross sections¹² for producing these nuclides. The ratio of saturation activities Sr⁹¹/Ba¹³⁹ was consistent with their production from uranium fission.¹² The Fielding bismuth in duplicate measurement showed an estimated 1.5-ppm (parts per million) impurity by weight (calculated as uranium).

The Berkeley group¹⁴ has measured formation cross sections for a number of nuclides produced in the

¹⁴ M. Lindner and R. N. Osborne, Phys. Rev. **94**, 1323 (1954); H. G. Hicks and R. S. Gilbert, Phys. Rev. **100**, 1286 (1955); P. C. Stevenson, H. G. Hicks, W. E. Nervik, and D. R. Nethaway, Phys. Rev. **111**, 886 (1958).

32-Mev proton fission of uranium. Extrapolating these data to the mass region 66–77, we find that the expected yields from a 1.5-ppm uranium impurity are smaller by factors of 10^3 – 10^5 than our observed yields. On the other hand, we do not consider it feasible to investigate with our present target material the possible complementary yield peak in the region of mass 130–140 because here the contribution from the uranium impurity would be of the same order of magnitude as the yields expected from bismuth, and hence must be accurately known.

The nuclides Ni^{66} , Cu^{67} , Zn^{71} , Ga^{72} , Ga^{73} , and As^{77} could be produced by spallation of zinc and germanium. Activation analyses for zinc in the Bi target were made by sandwiching the bismuth between ZnO layers deposited on aluminum and irradiating the stack with 14-Mev deuterons. From the average Zn^{69m} activity produced in the front and back ZnO layers (they differed by about 10%) and the Zn^{69m} activity found in the bismuth, the zinc concentration in the target was found in duplicate analyses to be 1.9 ± 0.3 ppm by weight.

Proton spallation of zinc could produce both Ni^{66} and Cu^{67} . A Zn target (3.1 mg/cm^2), electrodeposited on 1-mil Al, was irradiated with 36-Mev protons in a sandwich that included a Cu foil, an Al guard foil, the Zn target, and a second Al guard foil. The yields of Ni^{66} and Cu^{67} from zinc were measured after chemical separation from the irradiated target. Ni^{66} could be formed from the reactions $\text{Zn}^{68}(p,3p)$ and $\text{Zn}^{70}(p,\alpha p)$. Since our determination measures only the sum, weighted by the isotopic abundances of Zn^{68} and Zn^{70} in natural zinc, we report here only that the formation cross section of Ni^{66} from natural zinc is $4 \mu\text{b}$. Similarly, the formation cross section of Cu^{67} from natural zinc is 0.5 mb. The beam intensity as before was monitored by the $\text{Cu}^{65}(p,pn)\text{Cu}^{64}$ reaction in the Cu foil. Applying these cross sections to a 1.9-ppm zinc impurity in the bismuth results in negligible ($<0.1\%$) corrections for Ni^{66} and Cu^{67} . Thus even though the cross sections above are probably not known to better than 50%, the effect on the observed Ni^{66} and Cu^{67} yields can be disregarded.

The possibility of a substantial germanium impurity was excluded on indirect grounds. If germanium were present, one would expect a variety of arsenic activities from (p,xn) reactions. In fact, however, the decay curve and beta absorption curve of the arsenic fraction indicated only the presence of the 0.7-Mev beta of 39-hr As^{77} . Less than 5% of the observed activity could have come from arsenic isotopes of appreciably different half-life or beta energy. Thus we conclude that the germanium content is small and we can neglect spallation-produced contribution to the observed yields of Zn^{71} , Zn^{72} , Ga^{72} , Ga^{73} , and As^{77} .

We conclude then that the increased yield in the mass region 66–77 arises from bismuth fission and not from the activation of impurities.

IV. DISCUSSION

The 58-Mev mass distribution has the symmetric, sharply peaked shape similar to that observed by Fairhall¹ in 22-Mev deuteron fission of bismuth. Even at mass 67 where the fission yield is $5 \times 10^{-2}\%$, there is no evidence of appreciable departure from a symmetric type of fission.

The 36-Mev data indicate, however, that all of the yields in the mass region 66 to 77 are the same within less than a factor of two. The less probable yields of light products in a mass-yield distribution usually decrease exponentially with mass number. If we extrapolate with a slope determined by the yields at masses 77, 83, and 89, the yields at masses 66, 67, 72, and 73 should be (in microbarns) 0.026, 0.034, 0.17, and 0.23, respectively, if only symmetric fission is occurring. Our observed yields are much higher. Evidently asymmetric fission is also occurring.

The peak yield in the asymmetric mode occurs at about mass 70 (and presumably at mass 136 if $\bar{\nu}$, the average of neutrons per fission, is 4). The symmetric peak is at mass 103. In radium fission with 11-Mev protons,² the asymmetric peaks occur at about masses 87 and 135 and the symmetric peak at mass 111. In the fission of thorium, uranium, and heavier elements, the light peak moves to lower masses as the mass number of the target is reduced while the heavy peak remains essentially fixed. The same effect appears to exist here. The region of mass 130–140 will be investigated in the future with targets containing a lower uranium concentration.

If we sum the asymmetric peak, the cross section is about 6 microbarns or about 0.3% of the symmetric cross section (1900 microbarns). Thus the probability for the occurrence of asymmetric fission in bismuth at an excitation energy of about 41 Mev is very small.

Fairhall *et al.* have shown³ that the probability of fission for elements in the mass region of lead is very critically dependent on excitation energy. In fact Fairhall¹ concludes that the relative narrowness of the mass distribution in bismuth fission with 22-Mev deuterons is a consequence of the fission only of the compound nucleus Po^{211} . The fissionability of Po^{210} at ~ 10 Mev less excitation energy (i.e., following neutron evaporation) is very much less.

In this context we interpret the observed symmetric peak as follows. Since compound-nucleus formation is likely⁸ for 36-Mev protons on bismuth, the initial nucleus is Po^{210} excited to 41 Mev. From the fissionability curves¹⁵ of Fairhall *et al.* (expressed as σ_f/σ_R , the ratio of fission to total reaction cross section), we estimate σ_f/σ_R to be about 2×10^{-3} . Our observed σ_f ($\sim 99.7\%$ symmetric fission) is 1.9 mb; σ_R has been measured to be 1.3 b.⁸ Thus σ_f/σ_R is 1.5×10^{-3} , in reasonable agreement with the above. After evapora-

¹⁵ I. Halpern, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Palo Alto, California, 1959), Vol. 9, p. 245.

tion of one neutron, σ_f/σ_R is about 3×10^{-4} ; thus relatively little contribution from fission following neutron evaporation (Po^{209} , $E^* \sim 30$ Mev) is expected.

The origin of the asymmetric mode is more difficult to define within the Fairhall model,³ which suggests that the observed asymmetric fission must have occurred from an excitation energy of less than 20 Mev. The fissionability σ_f/σ_R at 20 Mev is very small ($\sim 4 \times 10^{-5}$ for Po^{208} and $\sim 4 \times 10^{-6}$ for Po^{210}).¹⁵ The observed asymmetric fission probability σ_a/σ_R is 5×10^{-6} . Thus if asymmetric fission of polonium nuclei is a low-energy phenomenon ($E^* < 20$ Mev), it appears to be competitive with symmetric fission, that is, $\sigma_a/\sigma_R \sim \sigma_f/\sigma_R$.

Fairhall and co-workers³ have made an empirical correlation of σ_a/σ_R with $B_n - E_{\text{th}} \pm \delta \equiv \Delta$ where B_n is the neutron binding energy, E_{th} is the fission threshold, and δ is a term that corrects for odd-even effects in both B_n and E_{th} .¹⁶ The correlation is intended to apply for relatively low excitation energies, $E^* \lesssim 12$ Mev. The ratio σ_a/σ_R appears to be a very sensitive function of Δ for $\Delta < 0$. Applied to our case, we must assume an average fissioning nucleus like Po^{207} in order to obtain sufficiently low E^* . For Po^{207} , B_n is 7.4 Mev¹⁷; δ is 0.7 Mev and is to be added for odd- A nuclei.¹⁶ Halpern¹⁵ estimates E_{th} from Fairhall's fission excitation functions to be ~ 18 Mev for Po nuclei. Thus Δ is about -10 Mev, in which case σ_a/σ_R is much less than 10^{-6} , according to our extrapolation of the empirical correlation. If the correlation is correct, one conclusion that can be drawn from these data is that the fission threshold in polonium nuclei is very large. An equation such as $E_{\text{th}} = 0.555(46 - Z^2/A)$ Mev, which appears to apply in the region of uranium,¹⁵ is clearly not applicable at polonium. From a different point of view, Halpern¹⁵ arrives at the same conclusion.

Although the shape of the asymmetric peak is not well defined by the data of this experiment, it seems inescapable that the peak is very narrow. The full width at half-height is only about 7 mass units, compared to 17 mass units for the symmetric peak and 15 mass units for the asymmetric peaks in 11-Mev proton fission² of Ra^{226} .

Fong¹⁸ has pointed out that the only very narrow peaks that have been observed in fission mass distributions are the fine-structure peaks in spontaneous fission and low-energy induced fission of thorium and some of the heavier elements.¹⁹ The fine structure, which occurs chiefly in the region of fragments with 82 neutrons and in the complementary region of light-mass

fragments, has been attributed^{18,20} to preferred formation of 82-neutron fragments in the fission act and enhanced post-fission neutron evaporation by products having somewhat more than 82 neutrons. These effects tend to disappear as the excitation energy is increased,²¹ there being little departure from a smooth curve in 14-Mev neutron-induced fission of U^{235} .

Noting that the complementary peak is near the 50-proton and 82-neutron shells, we suggest that the narrow peak observed in 36-Mev bismuth fission is a consequence of shell structure, as in the fine structure found in heavy-element fission. As Fairhall¹ has argued in discussing the narrow symmetric peak in 22-Mev deuteron fission of bismuth, we further attribute the narrowness of the asymmetric peak to the fission of a single nuclear species. If this species were of relatively low excitation energy (e.g., 20 Mev) at the time of fission, an asymmetric peak summing to a few microbarns should have been observed in Fairhall's experiment. Since the fission cross section with 22-Mev deuterons was only about $10 \mu\text{b}$ and appeared to be exclusively symmetric fission, we conclude that the excitation energy of the single species leading to the observed asymmetric fission was higher than 29 Mev (22 Mev plus deuteron binding energy), and indeed the most probable value may be 41 Mev.

As the excitation energy decreases from 40 Mev, $\sigma_f = \sigma_a + \sigma_s$ decreases by a factor of 10 in 10 Mev while σ_a/σ_s probably increases somewhat. Under these conditions the contribution to asymmetric fission of nuclei after the evaporation of one neutron must be small, and still smaller contributions are expected for fission after multiple neutron evaporation. At ~ 60 -Mev excitation, σ_f no longer decreases so rapidly with energy (a factor of 6 in 20 Mev). In this case multi-chance fission is likely, and the general broadening of the mass distribution as a consequence of the fission of several species increases the yields of products of large mass ratio. The lack of a separately resolved asymmetric mode with 58-Mev protons is consistent with these notions.

The suggestion that shell effects persist at relatively high excitation energies should be examined. In the case of 36-Mev protons forming a compound nucleus with Bi^{209} , the total excitation energy is about 41 Mev, of which perhaps 18 Mev goes to nuclear distortion and 23 Mev to nuclear excitation (less whatever rotational excitation there may be). According to Blatt and Weisskopf,²² a nucleus of mass 210 can be considered a simple degenerate gas to excitation energies of the order of 50 Mev. Most of the energy will be concentrated in those nucleons near the top of the potential well. In Po^{210} there are 78 nucleons before the $50p-82n$

¹⁶ J. D. Jackson, Atomic Energy of Canada Report AECL-329, 1956 (unpublished).

¹⁷ A. G. W. Cameron, Atomic Energy of Canada Report AECL-433, 1957 (unpublished).

¹⁸ Peter Fong (private communication, 1960).

¹⁹ L. E. Glendenin and E. P. Steinberg, *Annual Review of Nuclear Science* (Annual Reviews, Inc., Palo Alto, California, 1954), Vol. 4, p. 69.

²⁰ L. E. Glendenin, *Phys. Rev.* **75**, 337 (1949); L. E. Glendenin and C. D. Coryell, *Phys. Rev.* **77**, 755 (1950); D. R. Wiles, B. W. Smith, R. Horsley, and H. G. Thode, *Can. J. Phys.* **31**, 419 (1953).

²¹ A. C. Wahl, *Phys. Rev.* **99**, 730 (1955).

²² J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), p. 370.

shells are reached; an average excitation energy of 0.3 Mev per nucleon will take up all of the available energy, leaving these shells intact. Even if the probability that the excitation energy will be distributed in this way is small, this is still satisfactory for discussing a very improbable event like asymmetric fission. A value of 10^{-5} may be large enough.

On the other hand, in the case of a strongly deformed nucleus shell effects tend to disappear.²³ The situation for deformed nuclei excited to twenty-odd Mev is not clear. Nevertheless, it seems very likely that a closed-shell effect is important in almost all asymmetric fission since the heavy-mass peak is in the neighborhood of mass 135 for fissioning nuclei from californium to at least radium, and probably extending to polonium. For the purposes of this discussion, the closed shells need not be those found in spherical nuclei but may be some other configuration that is energetically favored.

On the basis of the present evidence we cannot rule out the possibility that asymmetric fission also occurs from nuclei of lower excitation energy, as a result of neutron evaporation following compound-nucleus formation or of an inelastic interaction of the proton with bismuth nuclei depositing only a fraction of the available momentum and energy. In this case we would expect from the arguments above that this contribution would be smaller (compared to that which was observed) and the distribution broader since several fissioning species would be involved. It is conceivable that the asymmetric peak broadens markedly at mass numbers below 66. Limitations due to the presence of trace impurities and the lack of adequate detection sensitivity prevent us from exploring these notions at this time.

V. ACKNOWLEDGMENTS

We are indebted to Dr. W. M. Preston, director of the Cyclotron Laboratory, for making this inter-university collaboration possible and to Mr. Richard Wharton for the many proton irradiations. Our thanks also to Professor N. S. Wall and Mr. Earle White for the deuteron bombardments at Massachusetts Institute of Technology. Mr. Taku Matsuo assisted in some of the measurements. We are particularly grateful to Professor Peter Fong for some illuminating discussions.

APPENDIX. CHEMICAL SEPARATION PROCEDURES

The Bi target was dissolved in ~ 10 ml of $6M$ HNO_3 . This solution was usually divided into aliquots ($\frac{1}{10}$ to $\frac{1}{2}$) and appropriate carriers added. In some cases several nuclides were isolated from the entire target solution.

²³ R. H. Lemmer and A. E. S. Green, Phys. Rev. **119**, 1043 (1960).

Substantially the same separation and purification procedures as those described in standard references^{24,25} were used for arsenic,²⁶ bromine,²⁷ strontium,²⁸ molybdenum,²⁹ and silver³⁰; they will not be discussed further here.

For elements whose nuclides are formed in very small yield (Ni, Cu, Zn, Ga) or that for which a method considerably different from the standard ones was used (Rb), a brief summary is given below. The choice of elements to be separated from a given aliquot as well as the size of the aliquot was a compromise among formation cross sections, half-lives, and chemical compatibility.

Nickel. Precipitated as Ni-dimethylglyoxime in ammoniacal solution in presence of excess citrate; scavenged with Bi-Cu-Pd-Sb sulfide, Pd-dimethylglyoxime, $BaCO_3-Fe(OH)_3$, and AgCl; purified by anion exchange on Dowex-1; counted as NiS.

Copper. Scavenged with $BaCO_3-Fe(OH)_3$ in ammoniacal solution, CdS (in presence of cyanide), and AgCl; purified by anion exchange on Dowex-1; counted as CuSCN.

Zinc. Precipitated as $ZnHg(SCN)_4$; scavenged with Bi_2S_3 and $BaCO_3-Fe(OH)_3$; purified by anion exchange on Dowex-1; counted as $ZnHg(SCN)_4$.

Gallium. Scavenged with Mo- α -benzoinoxime, Cd-Sb-Bi sulfide, $Fe(OH)_3$ (in $0.1M$ NaOH); extracted into diethyl ether; back-extracted into water and evaporated to dryness several times with HBr (to remove Ge, As, Se); extracted into diethyl ether; back-extracted with water; counted as Ga-8-hydroxyquinolate.

Rubidium. Coprecipitated with cesium as silicotungstates; scavenged with $BaCO_3-Fe(OH)_3$; Rb-Cs separated with $HBiI_4$; precipitated Bi_2S_3 ; precipitated $RbClO_4$; purified by cation exchange on Duolite C-3¹²; counted as $RbClO_4$.

Reagent blanks measured by processing an unirradiated Bi target through the chemical procedures gave values of <0.5 count/min for the nickel, copper, zinc, and arsenic fractions. These were measured on a counter whose background was 0.25 count/min.

²⁴ *Radiochemical Studies: The Fission Product*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951) National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, Part VI.

²⁵ "Collected radiochemical procedures," edited by J. Kleinberg, Los Alamos Report LA-1721 (rev.) 1954 (unpublished).

²⁶ R. J. Prestwood, reference 25, p. 91.

²⁷ L. E. Glendenin, R. R. Edwards, and H. Gest, reference 24, paper 232.

²⁸ L. E. Glendenin, reference 24, Paper No. 236.

²⁹ N. E. Ballou, reference 24, Paper No. 257.

³⁰ L. E. Glendenin, reference 25, p. 138.