Energy Dependence of the Recoil Properties of Products from the Interaction of U^{238} with 0.45-11.5-GeV Protons*

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The energy dependence of the integral recoil properties and formation cross sections of a number of barium and strontium nuclides produced in the interaction of U^{238} with protons has been studied between 0.45 and 11.5 GeV. The ranges and forward-to-backward ratios (F/B) of the neutron-excessive products are nearly independent of energy, and the results indicate that these products are formed in binary fission following interactions with low deposition energies. The ranges of the neutron-deficient products are practically constant up to 1 GeV, decrease by nearly a factor of 2 between 1 and 5 GeV, and decrease very slightly thereafter. The F/B values exhibit a pronounced peak in the vicinity of the midpoint of this interval. The results up to 1 GeV are consistent with binary fission. At this energy a different mechanism becomes noticeable and by 5 GeV this process predominates. Comparisons are made with the results expected for spallation, fission of a moderately light nucleus, and fragmentation. The latter is the only process which can account for the various experimental results. It is concluded that fragmentation is the main mechanism for the formation of neutron-deficient products lying in the "fission" region for incident energies above 5 GeV.

I. INTRODUCTION

The mechanism for the formation of neutron-deficient nuclides lying in the fission region from the interaction of uranium with GeV protons has been of interest for some time. The first indications that processes other than binary fission, which leads to the formation of these nuclides below 1 GeV, are of importance were given by Friedlander *et al.*[1] and Alexander, Baltzinger, and Gadzik.[2] The first of these groups found that the charge dispersion in the 125–140 mass region changed from the single-humped curve normally associated with binary fission to a double-humped curve at a bombarding energy somewhat below 1 GeV. Alexander, Baltzinger, and Gadzik[2] determined that at 6 GeV the recoil ranges of neutron-deficient iodine isotopes were nearly a factor of 2 smaller than those of neutron-excessive iodines whereas there was practically no difference between these ranges at 0.7 GeV. Furthermore, the deposition energies associated with the formation of these neutron-deficient nuclides were unusually low at 6 GeV and actually appeared to decrease with increasing bombarding energy. The isotopic cross sections also showed a marked change between 0.7 and 6 GeV, not inconsistent with the results of Friedlander *et al.*[1]

More recent studies have given additional evidence of a change in mechanism at GeV energies. Rudstam and Sørensen[3] measured the cross sections of a large number of iodine nuclides at 0.59 and 18 GeV. Their isotopic-yield distribution at 18 GeV suggested that products with $A \leq 122$ were primarily formed by a spallation process, while those with $A > 123$ were the result of binary fission. Brandt[4] determined the ranges of these same nuclides at the above energies. He finds that at 0.59 GeV the ranges of all the iodine isotopes are nearly identical and their magnitude is characteristic of fission. On the other hand, at 18 GeV the ranges of very neutron-deficient products are only half as large as those of the fission products, with a sharp transition occurring at $A \sim 123$. Similar isotopic-yield[5] and recoil results[6] have also been reported for antimony isotopes. Isobaric-yield distributions at GeV energies have recently been reported for $A = 109$,[7] 111,[8] and 117,[9] and earlier measurements at 2.9 and 28 GeV have been summarized in a recent review.[10] These charge-dispersion results are consistent with a double-peaked distribution although it appears that the peaks overlap to a substantial extent. A twin-peaked charge-dispersion curve has also been reported for rare-earth products from uranium at 28 GeV.[11,12] Recoil measurements of isobaric products in the $A \sim 111$ mass region have recently been reported for both GeV[8,9] and lower energies.[9,13] The results of these studies are qualitatively similar to those described above for iodine products. More detailed information about the recoil properties of neutron-deficient products has been given in a recent report[14] of their differential ranges and angular distributions at 2.2 GeV. The authors find the data to be consistent with fission of a nucleus with $A \sim 203$ although a small spallation contribution is necessary to account for the lowest-energy fragments.

In spite of this growing body of experimental information about the properties of the neutron-de-

ficient products, the mechanism by which they are produced has not as yet been definitively established. The experiments cited above have been interpreted in terms of spallation, fission, fragmentation, or a combination of these processes. In order to obtain more precise information about which of these mechanisms is primarily responsible for the observed results we have measured the energy dependence of the recoil properties, as well as the excitation functions, of several neutron-deficient products formed in the interaction of U^{238} with 0.45–11.5-GeV protons. The above processes should, at least in principle, lead to a significantly different energy dependence of the recoil properties. As shown in Sec. IV, spallation should lead to a decrease in range at a substantially lower bombarding energy for a heavy product such as Ba^{131} than for a light product such as Sr^{83}. A fission mechanism ought to lead to a gradual decrease in range over a wide bombarding-energy region as the mass of the fissioning nucleus decreases. One would presumably expect a similar behavior for Ba^{131} and Sr^{83}. Fragmentation would require, in addition to the observed decrease in range, a concomitant breakdown of the two-step model as manifested, for instance, by anomalously low deposition energies.

The experimental procedure is described in Sec. II and the results for Sr^{82}, Sr^{83}, Ba^{128}, Ba^{131}, as well as for neutron-excessive Sr^{91} and Ba^{140} are presented in Sec. III. A detailed comparison with the results expected for the above processes is given in Sec. IV.

II. EXPERIMENTAL PROCEDURE

The thick-target–thick-catcher technique[15] was used to determine the average range in uranium of the recoil products. Targets consisted of 0.001- or 0.002-in.-thick depleted uranium foils sandwiched between 0.001-in.-thick aluminum of high (99.999%) purity. The aluminum foils were slightly larger than the target foil in order to insure the collection of recoils escaping from the edges. In some of the early experiments an additional aluminum foil was included in the target stack for determination of barium and strontium activities from impurities in the aluminum. This effect was found to be completely negligible. The target stack was wrapped with another high-purity aluminum foil and then mounted on a target holder for irradiation. Just prior to target assembly, the uranium foil was washed with 6 N HNO_3 to remove the oxide layer.

For experiments designed to measure the cross sections of the products of interest the target and catcher foils were carefully cut to the same size. Additional aluminum foils, used to monitor the beam intensity by means of the $Al^{27}(p, 3pn)$ reaction, were placed on either side of the recoil catchers and the whole assembly was wrapped with this same material. After irradiation the target stack was carefully trimmed on all sides to minimize errors resulting from possible misalignment of the various foils.

Irradiations were performed with 0.45-, 1.0-, 1.5-, 2.0-, 3.0-, 4.5-, 6.0-, 9.0-, and 11.5-GeV protons. The bombardments at 0.45 GeV were performed in the circulating beam of the University of Chicago synchrocyclotron and those at 1 GeV in that of the Princeton-Pennsylvania accelerator. All other irradiations were performed in the internal beam of the zero-gradient synchrotron at Argonne National Laboratory. In all cases the targets were oriented at 90° to the beam direction.

Following bombardment the target and catcher foils were separately dissolved in acid and strontium and barium radiochemically separated. In the case of the cross-section measurements, the target and catcher foils were dissolved together in order to obtain the total yield. Standard[16] chemical procedures were used, involving primarily precipitations with fuming HNO_3 and with Na_2CrO_4. In some of the early experiments barium was also

TABLE I. Decay properties of observed radionuclides.[a]

Nuclide	Half-life	Radiation	Branching ratio (%)
$Sr^{82}(Rb^{82})$[b]	25.0 day	β^+	96
Sr^{83}	33.0 h	β^+	16
$Sr^{91}(Y^{91m})$[b]	9.7 h	0.55-MeV γ	63
$Ba^{128}(Cs^{128})$[b]	2.4 day	β^+	70
Ba^{131}	11.6 day	0.216-MeV γ	20
Ba^{140}	12.8 day	0.537-MeV γ	25
$Ba^{140}(La^{140})$[b]	12.8 day	1.596-MeV γ	96
Na^{24}	15.0 h	1.360-MeV γ	100

[a] Based on properties summarized by C. M. Lederer, J. M. Hollander, and I. Perlman, in *Table of Isotopes* (John Wiley & Sons, New York, 1967), 6th ed., and on the following more recent reports: Ba^{128}: T. Kucarova, V. Z. Zvolska, B. Kracik, and A. Mastalka, Czech. J. Phys. 18, 24 (1968); Ba^{131}: T. Kucarova, B. Kracik, and V. Z. Zvolska, Yadern. Fiz. 7, 713 (1968) [transl.: Soviet J. Nucl. Phys. 7, 433 (1968)]; Ba^{140}: J. Kern and G. Mauron, Helv. Phys. Acta. 43, 272 (1970); Sr^{83}: R. C. Etherton, L. M. Beyer, W. H. Kelly, and D. J. Horen, Phys. Rev. 168, 1249 (1968); A. S. Basina *et al.*, Izv. Akad. Nauk SSSR, Ser. Fiz. 32, 1633 (1968) [transl.: Bull. Acad. Sci. USSR, Phys. Ser. 32, 1518 (1968)]; Sr^{91}: J. D. Knight, O. E. Johnson, A. B. Tucker, and J. E. Solecki, Nucl. Phys. A130, 433 (1969).

[b] Radiation detected is that of daughter in equilibrium with parent of interest.

TABLE II. Recoil properties of observed nuclides.

	Ba^{140}		Ba^{131}			Ba^{128}		
E_p (GeV)	$2W(F+B)$ (mg/cm^2)	F/B	$2W(F+B)$ (mg/cm^2)	F/B	No. of determinations	$2W(F+B)$ (mg/cm^2)	F/B	No. of determinations
0.45	8.18±0.15	1.06±0.01	7.39±0.23	1.37±0.03	3	6.35±0.30	1.86±0.30	1
1.0	7.80±0.50	1.03±0.06	7.21±0.47	1.37±0.08	2			
1.5	8.18±0.13	1.07±0.03	6.58±0.18	1.44±0.08	3	6.42±0.43	1.67±0.06	2
2.0	7.87±0.27	1.06±0.04	5.14±0.35	1.55±0.02	5	5.79±0.17	1.50±0.16	2
3.0	7.27±0.31	1.08±0.08	4.46±0.16	1.66±0.01	3	4.15±0.20	1.80±0.20	1
4.5	7.55±0.05	1.11±0.03	3.85±0.10	1.59±0.03	2	3.42±0.07	1.74±0.06	2
6.0	8.06±0.18	1.07±0.05	3.92±0.18	1.41±0.08	4	3.34±0.15	1.50±0.10	1
9.0	8.03±0.24	1.03±0.03	3.76±0.11	1.31±0.04	2	3.10±0.10	1.34±0.02	2
11.5	7.95±0.26	1.04±0.03	3.58±0.11	1.27±0.02	3	2.99±0.15	1.25±0.15	1
	Sr^{91}		Sr^{83}			Sr^{82}		
0.45	10.70±0.20	1.06±0.02	10.02±0.07	1.31±0.06	2	9.51±0.30	1.35±0.20	1
1.0	10.75±0.35	1.06±0.05	10.24±0.15	1.38±0.20	2			
1.5	10.50±0.25	1.13±0.05	9.62±0.25	1.48±0.04	3	7.95±0.80	1.78±0.55	4
2.0	10.11±0.31	1.09±0.04	8.57±0.30	1.44±0.06	5	8.21±0.68	1.41±0.19	5
3.0	10.22±0.25	1.03±0.02	8.04±0.16	1.32±0.06	2	7.69±0.31	1.43±0.06	3
4.5	9.94±0.25	1.12±0.03	6.77±0.23	1.35±0.03	2	6.14±0.52	1.47±0.07	2
6.0	10.23±0.35	1.08±0.04	6.35±0.15	1.18±0.02	4	6.33±0.12	1.24±0.01	2
9.0	10.00±0.15	1.09±0.04	6.20±0.15	1.12±0.03	2	5.24±0.10	1.11±0.01	2
11.5	10.09±0.13	1.09±0.04	6.12±0.25	1.18±0.08	4	5.35±0.20	1.06±0.15	1

separated from radium by ion exchange using a procedure described elsewhere.[17] Since it was determined that at least some of the prominent Ba^{131} γ rays were completely free of radium interference, this step was discontinued.

The activity measurements were based on the detection of either γ rays or positrons. The γ-ray measurements were performed with a calibrated Ge(Li) detector used in conjunction with a 1024-channel pulse-height analyzer. The positron measurements were based on detection of the coincident annihilation quanta with two NaI(Tl) detectors. The efficiency of this system was determined with a standardized Na^{22} source. The activity of Na^{24} from the $Al^{27}(p, 3pn)$ monitor reaction was directly assayed with the Ge(Li) detector. The upstream monitor foil was used for this purpose, while the downstream foil served as a check on the alignment procedure. The Na^{24} activity of the latter was usually 1–2% higher than that of the former as a result of secondary reactions. In one instance the difference between the monitor foils was substantially larger, presumably due to misalignment. The results of this experiment were discarded. Table I summarizes the relevant information concerning the nuclides determined in this study, including their half-lives, measured radiations, and assumed abundances.

III. RESULTS

A. Recoil Properties and Cross Sections

The quantities determined in the recoil experiments are the fraction of the total activity of a given nuclide collected in the forward and backward catchers, denoted by F and B, respectively. The recoil properties of interest are the experimental range, $2W(F+B)$, and the forward-to-backward ratio, F/B. The target thickness is W mg/cm^2. The ranges have been corrected for scattering[18] (4%) and edge[19] (0.5–2%) effects. The

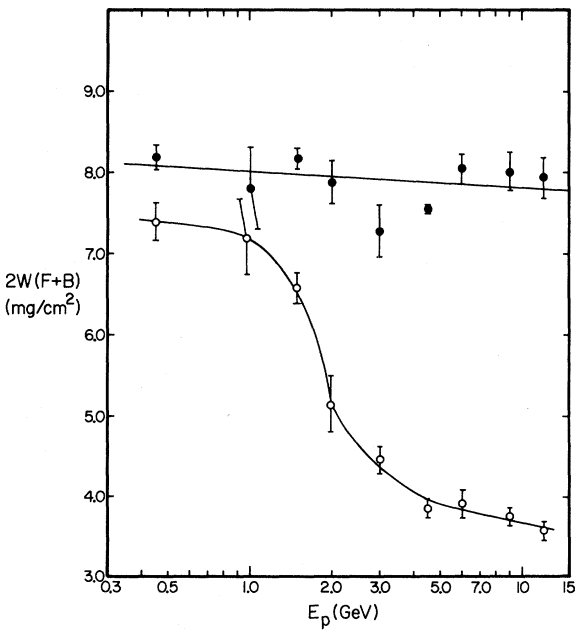

FIG. 1. Energy dependence of the experimental ranges $[2W(F+B)]$ of Ba^{131} (o) and Ba^{140} (●).

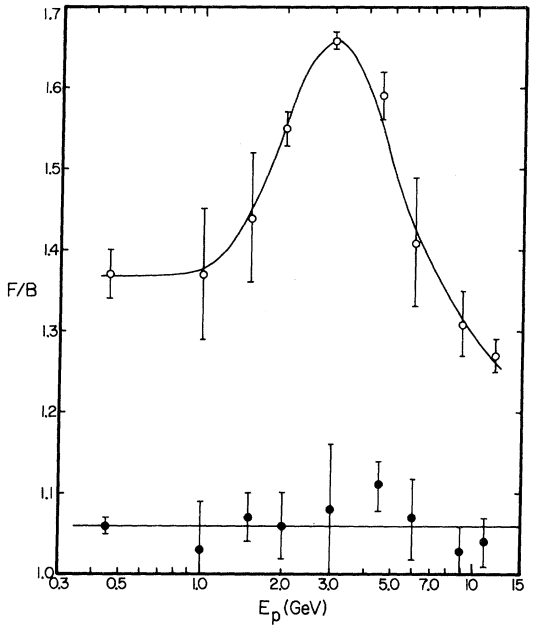

FIG. 2. Energy dependence of the forward-to-backward ratios of Ba^{131} (o) and Ba^{140} (●).

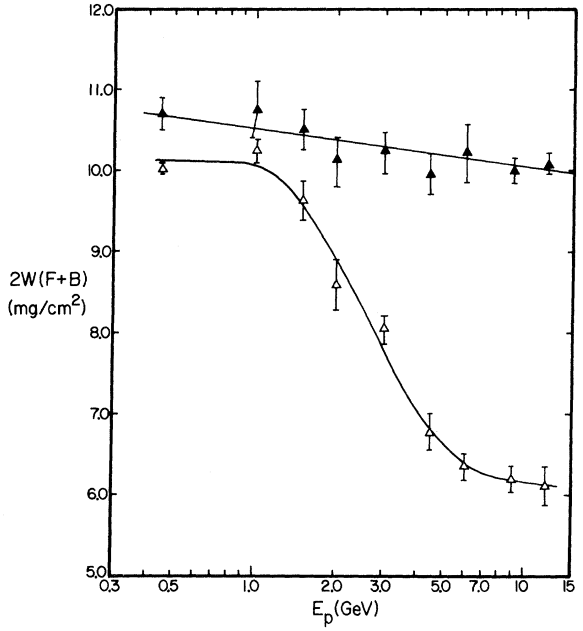

FIG. 3. Energy dependence of the experimental ranges $[2W(F+B)]$ of Sr^{83} (Δ) and Sr^{91} (▲).

corrected recoil properties are summarized in Table II. Usually 2 to 5 separate determinations were made at each energy and the average values are listed. The quoted errors are experimental standard deviations. The results for Ba^{128} and Sr^{82} are less complete and in those cases where only a single measurement was made an estimate of the error is given.

The energy dependence of the measured recoil properties is shown in Figs. 1 and 2 for Ba^{131} and Ba^{140} and in Figs. 3 and 4 for Sr^{83} and Sr^{91}. The results for Sr^{82} and Ba^{128} are qualitatively similar to those for the other neutron-deficient isotopes, albeit of lesser accuracy. Whereas the ranges of the neutron-excessive products are nearly independent of bombarding energy, decreasing by less than 10% between 0.45 and 11.5 GeV, those of the neutron-deficient products have a completely different energy dependence. The ranges of these nuclides are practically independent of energy up to 1 GeV, decrease by nearly a factor of 2 between 1 and 5 GeV, and decrease only slightly thereafter. The midpoint of this drop occurs at about 1.9 GeV for Ba^{131} and 2.5 GeV for Sr^{83} but the uncertainties are such that this difference is probably not too significant.

The F/B values of the neutron-deficient products display a totally unexpected energy dependence. The values initially increase with energy, peak at 2–3 GeV, and decrease rapidly thereafter. By contrast, the F/B of the neutron-excessive products are essentially independent of energy and are only slightly larger than unity.

The formation cross sections were determined relative to that of the $Al^{27}(p, 3pn)$ reaction. The values of the latter were taken from the review article by Cumming.[20] The results for the neu-

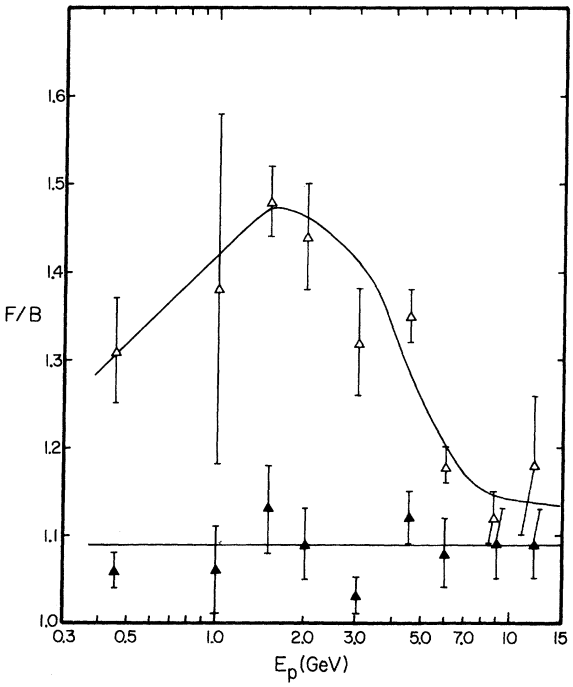

FIG. 4. Energy dependence of the forward-to-backward ratios of Sr^{83} (Δ) and Sr^{91} (▲).

TABLE III. Formation cross sections of observed products (in mb).

E_p (GeV)	Ba^{140}	Ba^{131}	Ba^{128}	Sr^{91}	Sr^{83}	Sr^{82}
0.45	12.1±0.2	3.2±0.1	...	27.5±0.3	0.10±0.01	0.013±0.004
1.5	8.3±1.0	6.7±1.0	2.7±0.4	21.4±2.0	...	...
2.0	7.8±0.3	6.9±0.3	3.4±0.1	16.5±0.5	0.59±0.01	0.082±0.001
3.0	7.4±0.2	8.2±0.2	5.1±0.1	14.2±0.4	4.1 ±0.1	0.74 ±0.05
4.5	7.5±0.3	9.6±0.3	6.0±0.1	15.0±0.7	5.0 ±0.3	1.1 ±0.1
6.0	7.6±0.1	9.4±0.1	6.6±0.8	14.7±0.2	7.1 ±0.3	1.3 ±0.1
9.0	7.4±0.2	8.4±0.1	5.6±0.1	13.4±0.6	5.6 ±0.2	1.3 ±0.1
11.5	7.8±0.2	8.8±0.1	6.5±0.5	14.3±0.4	5.8 ±0.2	1.4 ±0.1

tron-deficient products were increased by 4% to correct for the effect[21] of secondary reactions on the cross section of the monitor reaction. The cross sections of the neutron-excessive products are presumably affected by secondary reactions to a comparable extent as the monitor reaction, and the results were consequently not corrected for this effect. The results are summarized in Table III where all the listed values represent cumulative yields. The excitation functions for the production of Sr^{83} and Ba^{131} are plotted in the top panel of Fig. 5. Acting on the assumption that the energy dependence of the recoil ranges is indicative of a transition from fission to an as yet unspecified high-energy mechanism, it is possible to decompose the excitation functions into curves associated with these individual processes. We assume that only fission contributes below 1 GeV, and that the fission range has the same energy dependence as the ranges of the neutron-excessive products. It is assumed that the range associated with the high-energy mechanism is independent of energy and has a value that is 1 standard deviation smaller than the measured value at 11.5 GeV. The relative contribution of the two mechanisms is easily determined on the basis of these assumptions, and the excitation functions associated with the high-energy mechanism are shown in the bottom panel of Fig. 5. These curves must, of course, be regarded as highly approximate. Nonetheless, they do indicate that the cross sections associated with the high-energy process increase rapidly up to approximately 6 GeV and then level off or perhaps even decrease.

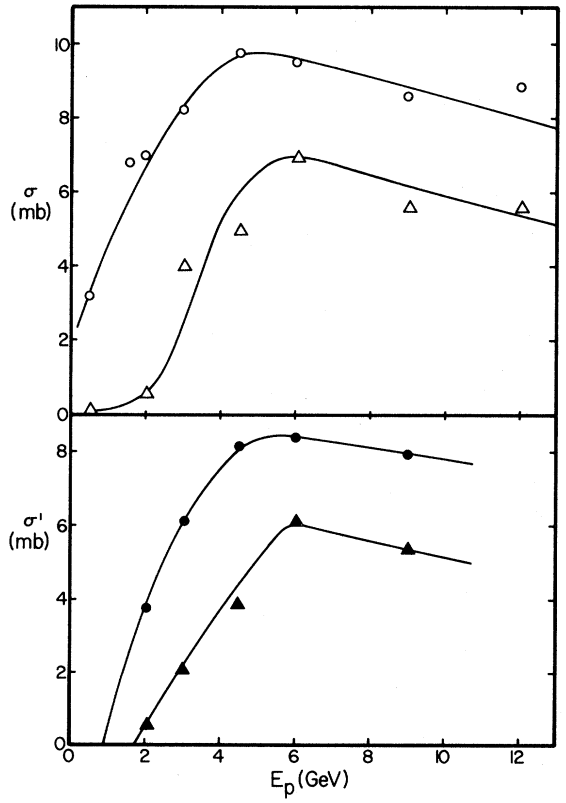

FIG. 5. Top panel – excitation functions for the formation of Ba^{131} (o) and Sr^{83} (Δ); bottom panel – exictation functions for the formation of Ba^{131} (●) and Sr^{83} (▲) by the high-energy mechanism.

The results of the cross-section and recoil-property measurements may be compared with previous determinations. Sugarman and collaborators[18,22] measured the recoil properties of a large number of nuclides at 0.45 GeV, including Ba^{140} and Sr^{91}. Friedlander *et al.*[1] determined the excitation functions of Ba^{140} and Ba^{131} and also measured their recoil properties at 2.9 GeV. Table IV presents a comparison with these results. The agreement is generally quite good except for a discrepancy with the ranges determined by Friedlander *et al.*,[1] which are substantially larger than the present values.

B. Recoil Parameters

The recoil properties may be used to derive the values of the various recoil parameters by means of the velocity vector model[23,15,18] embodying the two-step mechanism commonly invoked in high-en-

TABLE IV. Comparison with previous measurements.

Product	Energy (GeV)	Recoil properties: Previous measurement $2W(F+B)$ (mg/cm^2)	F/B	Reference	Present result $2W(F+B)$ (mg/cm^2)	F/B
Ba^{140}	0.45	7.96±0.02	1.10±0.04	18	8.18±0.15	1.06±0.01
		7.98±0.07	1.10±0.04	22		
	2.9	8.02±0.40 [a]	1.09±0.05	1	7.27±0.31	1.08±0.08
Ba^{131}	2.9	5.60±0.30 [a]	1.89±0.10	1	4.46±0.16	1.66±0.01
Sr^{91}	0.45	10.45±0.08	1.13±0.04	18	10.70±0.20	1.06±0.02
		Cross sections (mb)			(mb)	
Ba^{140}	2.9	7.9 ±0.2		1	7.4 ±0.2	
Ba^{131}	2.0	6.8 ±0.2		1	6.9 ±0.3	
	2.9	8.55±0.05		1	8.2 ±0.2	
	6.2	6.6 ±0.9		1	9.4 ±0.1	

[a]The quoted values were reduced by 5% to correct for scattering and edge effects.

ergy reactions. Let the forward component of the average impact velocity of the struck nucleus be denoted by $v_\parallel$ and the average velocity associated with the deexcitation step by V. The ratio of these velocities is given by the quantity $\eta_\parallel$. The range R_0, corresponding to the velocity V, and $\eta_\parallel$ are obtained from the measured recoil properties by the relations:

$$2W(F+B) = R_0[1 + \tfrac{1}{4}\eta_\parallel^2(N+1)^2], \quad (1)$$

$$\frac{F}{B} = \frac{1 + \frac{2}{3}\eta_\parallel(N+2) + \frac{1}{4}\eta_\parallel^2(N+1)^2}{1 - \frac{2}{3}\eta_\parallel(N+2) + \frac{1}{4}\eta_\parallel^2(N+1)^2}. \quad (2)$$

The quantity N is the exponent in the relation between range and velocity in the lab system. These equations represent the first terms of an expansion and are only accurate when $\eta_\parallel \ll 1$ not only on the average, but also for each interaction. The effect of broad and overlapping distributions of $v_\parallel$ and V has been considered in detail by Porile[24] and will be discussed below. These formulas also assume that the angular distribution of the breakup velocity V in the moving frame is isotropic and that the transverse component of the impact velocity, $v_\perp$, is zero. These assumptions introduce a minor error in the values of R_0 and $\eta_\parallel$. For instance the assumption that $v_\perp/v_\parallel = 7$, a value that is suggested by a calculation described in Sec. IV C, increases $\eta_\parallel$ calculated for Ba^{131} at 11.5 GeV by 4% and decreases R_0 by a similar percentage. This estimate is based on more complete forms of Eqs. (1) and (2) that include a term in $\eta_\perp^2$.

The average kinetic energies of the products, denoted by T, were obtained from the corresponding R_0 by means of a range-energy relation discussed in detail elsewhere.[24] This relation is based on the energy losses of heavy ions in UF_4 determined by Bridwell and Moak[25] coupled with the ranges of products from the fission of U^{235} by thermal neutrons determined by Niday.[26] The range-energy relation depends on fragment Z and, since the isolated products include the contributions of all their isobaric progenitors, effective Z values had to be determined. This was done on the basis of previous charge-dispersion measurements.[1,8,27] The range-energy relation in question was also used to determine for each product the value of the constant N appearing in Eqs. (1) and (2). Since N, in fact, varies slowly with the kinetic energy of the fragment, this determination was made at the average value of T.

The values of T and $\eta_\parallel$ may, in turn, be used to determine the average values of $v_\parallel$. This quantity has been related[15,28] to the average deposition energy E^* of the residual nuclei leading to the products in question. The relation

$$E^* = 0.80E_p M_R v_\parallel / P_p \quad (3)$$

relating E^* to $v_\parallel$, the momentum P_p and kinetic energy E_p of the incident proton, and the mass of the residual nucleus following the cascade, M_R, has been derived from Monte Carlo cascade calculations performed up to 1.8 GeV.[29] In the absence of calculations at higher energies, Eq. (3) has been applied over the entire energy range covered in this experiment. In all instances M_R was taken as 235 amu.

The differential ranges of the neutron-deficient products of interest are known[14] to be broad at GeV energies. The errors introduced in the vector-model analysis of average recoil properties associated with broad-range curves have been in-

vestigated elsewhere.[24] The principal sources of error turn out to be the overlap between the $v_{\parallel}$ and V distributions and the use of a constant value of N to represent a quantity that is, in fact, velocity dependent. It has been estimated[24] that the vector-model analysis of Ba^{131} recoil properties at 6–12 GeV overestimates the value of V by some 5% and underestimates the value of $v_{\parallel}$, and hence also that of E^*, by as much as 25%. It should also be noted, however, that E^* is proportional to the assumed mass of the residual nucleus. The adopted value of 235 amu is undoubtedly much too large for neutron-deficient products far removed from the target. The formation of these products requires large deposition energies regardless of the mechanism involved, and cascade calculations indicate[29] that such interactions involve the emission of a sizable number of cascade nucleons. These two errors in the calculation of E^* values for neutron-deficient products will thus, in large measure, cancel each other.

The various recoil parameters derived from the measured recoil properties in the manner described above are summarized in Table V. The

TABLE V. Recoil parameters derived from measured properties.

E_p (GeV)	R_0 (mg/cm^2)	$\eta_{\parallel}$	T (MeV)	$v_{\parallel}$ (MeV/amu)$^{1/2}$	E^* (MeV)
		Ba^{131}			
0.45	7.34±0.23	0.0652±0.0045	49.5±2.1	0.0566±0.0040	143±10
1.0	7.16±0.47	0.0652±0.0119	47.9±4.1	0.0557±0.0104	188±35
1.5	6.52±0.18	0.0755±0.0112	42.4±1.5	0.0607±0.0091	232±35
2.0	5.07±0.35	0.0906±0.0026	31.2±2.5	0.0626±0.0031	258±13
3.0	4.38±0.16	0.1047±0.0012	26.5±1.0	0.0666±0.0015	300±7
4.5	3.79±0.10	0.0869±0.0034	22.6±0.6	0.0510±0.0021	246±10
6.0	3.88±0.18	0.0644±0.0104	23.2±1.1	0.0383±0.0063	192±31
9.0	3.74±0.11	0.0506±0.0057	22.3±0.7	0.0295±0.0033	154±17
11.5	3.56±0.11	0.0448±0.0029	21.3±0.6	0.0255±0.0017	136±9
		Ba^{140}			
0.45	8.18±0.15	0.0127±0.0020	61.8±1.6	0.0119±0.0019	30±5
1.0	7.80±0.50	0.0064±0.0127	57.9±5.0	0.0058±0.0115	20±39
1.5	8.18±0.13	0.0147±0.0061	61.8±1.4	0.0138±0.0057	53±22
2.0	7.87±0.27	0.0127±0.0082	58.6±2.7	0.0116±0.0075	48±31
3.0	7.27±0.31	0.0167±0.0161	52.7±3.0	0.0145±0.0140	65±63
4.5	7.54±0.05	0.0227±0.0059	55.4±0.5	0.0202±0.0052	97±25
6.0	8.06±0.18	0.0147±0.0102	60.5±1.9	0.0137±0.0094	68±47
9.0	8.03±0.24	0.0064±0.0063	60.3±2.5	0.0060±0.0059	31±31
11.5	7.95±0.26	0.0085±0.0063	59.4±2.6	0.0079±0.0058	42±31
		Sr^{83}			
0.45	9.97±0.07	0.0583±0.0098	77.6±0.8	0.0798±0.0134	201±34
1.0	10.17±0.15	0.0695±0.0308	79.8±1.6	0.0964±0.0426	326±144
1.5	9.52±0.25	0.0846±0.0057	72.8±2.6	0.1120±0.0078	428±30
2.0	8.49±0.30	0.0787±0.0088	62.4±2.9	0.0965±0.0110	398±45
3.0	8.00±0.16	0.0600±0.0097	57.7±1.5	0.0707±0.0115	318±52
4.5	6.73±0.23	0.0648±0.0047	46.5±1.9	0.0686±0.0052	331±25
6.0	6.34±0.15	0.0322±0.0033	42.7±1.2	0.0326±0.0034	163±17
9.0	6.19±0.15	0.0220±0.0052	41.6±1.2	0.0220±0.0052	115±27
11.5	6.11±0.25	0.0322±0.0131	40.9±2.0	0.0319±0.0130	170±69
		Sr^{91}			
0.45	10.70±0.20	0.0127±0.0041	94.5±2.5	0.0183±0.0059	46±15
1.0	10.75±0.35	0.0127±0.0102	95.2±4.4	0.0183±0.0148	62±50
1.5	10.49±0.25	0.0266±0.0096	91.9±3.1	0.0378±0.0137	144±52
2.0	10.10±0.31	0.0187±0.0080	87.2±3.7	0.0259±0.0110	107±46
3.0	10.22±0.25	0.0064±0.0042	88.6±3.0	0.0090±0.0059	40±27
4.5	9.93±0.25	0.0246±0.0058	85.2±3.0	0.0337±0.0080	162±38
6.0	10.23±0.35	0.0167±0.0080	88.7±4.3	0.0234±0.0112	117±56
9.0	9.99±0.15	0.0187±0.0080	85.9±1.8	0.0257±0.0110	134±57
11.5	10.08±0.13	0.0187±0.0080	87.0±1.6	0.0259±0.0110	138±59

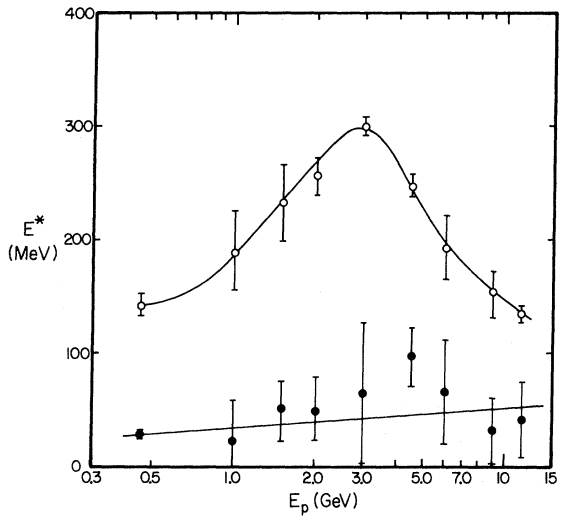

FIG. 6. Energy dependence of the average deposition energy of residual nuclei leading to Ba^{131} (o) and Ba^{140} (●).

listed uncertainties are based solely on those of the recoil properties. Of special interest for an understanding of the reaction mechanisms are the deposition energies, plotted in Figs. 6 and 7. The E^* values associated with the formation of the neutron-excessive products are low and increase only slightly with energy. These products are clearly formed in the same type of interaction at all bombarding energies. The decreasing yield of these products with energy indicates that low-energy transfers leading to fission slowly become less probable as the proton energy increases.

The deposition energies associated with the formation of the neutron-deficient products initially increase with bombarding energy. This is the behavior expected for products that require large energy transfers for their formation. The increase in E^* merely reflects the increasing contribution of high values to the excitation energy spectrum of the residual nuclei. According to this viewpoint[30] one might expect the E^* values to increase until the probability of transferring the excitation energy required to form the product no longer increases with proton energy, and then to level off. The results, however, do not bear out this expectation and show that the E^* values decrease above 2–3 GeV. This finding is consistent with previous[2,6,8,9] determinations at 6, 12, and 18 GeV which showed that the E^* values of various neutron-deficient products from uranium were lower at these energies than at 0.4–0.6 GeV. The observed decrease must surely result from a breakdown of one or more of the assumptions of the velocity vector model. A real decrease of deposition energies, coupled with the still rising excitation functions, would be impossible to explain on the basis of any plausible mechanism.

IV. DISCUSSION

In this section the experimental results will be analyzed in terms of the three mechanisms that have been postulated for the production of the neutron-deficient products at multi-GeV energies: fission, spallation, and fragmentation. We consider the three principal results requiring explanation to be: (1) the low ranges and forward-to-backward ratios obtained above 5 GeV; (2) the nearly identical sharp decrease of the ranges of products of widely differing masses between 1 and 4 GeV; (3) the decrease in E^* values above 2–3 GeV and the apparent correlation of this decrease with that of the ranges. The results obtained up to 1 GeV require no further comment, since a binary-fission process is clearly indicated. This is also the case for the neutron-excessive products at all energies.

A. Fission

The possibility that binary fission remains the principal mechanism for the formation of the neutron-deficient products at GeV energies has been considered by various groups.[1,8,14] In view of the low ranges of these products it is obvious that the average fissioning nuclei must be considerably lighter than those leading to the formation of the

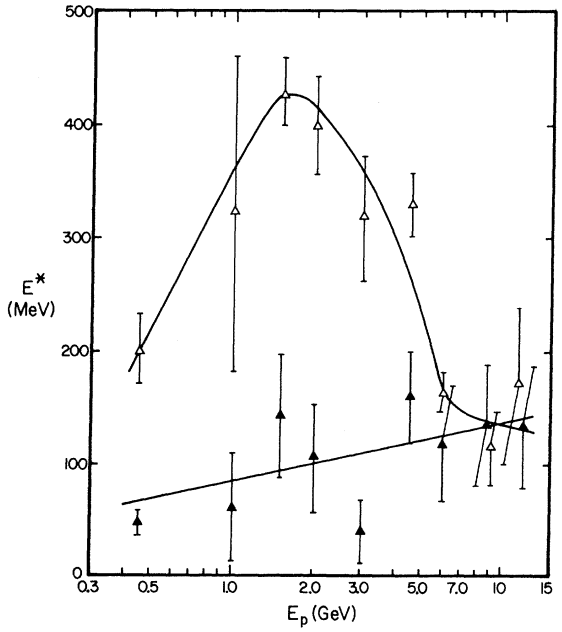

FIG. 7. Energy dependence of the average deposition energy of residual nuclei leading to Sr^{83} (△) and Sr^{91} (▲).

neutron-excessive products. Let us estimate the mass numbers A_F of the average fissioning nuclei for different bombarding energies and see if they are physically plausible.

An estimate of the identity of the fissioning nuclei can be made by comparing the measured ranges with those of the same products formed in the interaction of nuclei lighter than uranium with protons of a sufficiently low energy to ensure that fission is still the principal mechanism. This procedure assumes that there is a unique relation between the range of a fragment and the identity of its fissioning nucleus irrespective of bombarding energy or target. This assumption is not as sweeping as it might appear, since fission is a slow process on the time scale of high-energy interactions. A given fissioning nucleus should thus give rise to fragments having the same kinetic energies regardless of its mode of formation, since it retains no memory of this event. The inverse statement, that fragments having a given range or energy result from the same average fissioning nucleus, whatever the target or bombarding energy might be, is perhaps less secure. Its validity depends on the subsidiary requirement that the charge-to-mass ratio of fissioning nuclei of a given mass be independent of the system under consideration. The similarity of the distribution of spallation products resulting from different targets and energies exemplified, for instance, by the isotopic yields of iodine nuclides from a variety of targets at 0.6 and 18 GeV,[3] lends credence to this assumption.

The recoil properties of various strontium and barium nuclides have been determined for the fission of bismuth and tantalum by 0.45-GeV protons.[15] These data, coupled with the present results at this energy for uranium form the basis of our estimate. The ranges reported[15] for Sr^{91}, Ba^{128}, and Ba^{133m} were corrected for target thickness and scattering[19] and then converted to ranges in uranium using empirically determined conversion factors reported elsewhere.[19] The ranges of Sr^{83} and Ba^{131} were inferred from the measured values for these neighboring isotopes on the basis of the variation of range with mass number obtained for strontium and barium nuclides at 0.45 GeV in the present work. Small corrections for the difference between the actual and effective charge resulting from the cumulative nature of the measured yields were first made on the basis of known[1,18,31] charge dispersions. The identity of the average fissioning nuclei leading to the products of interest has been inferred[15] from the 0.45-GeV recoil data for bismuth and tantalum as Pb^{191} and Hf^{164}, respectively. U^{234} was similarly[22] chosen as the average fissioning nucleus for uranium at 0.45 GeV. Smooth curves were then drawn through the three (R_0, A_F) points for Sr^{83} and Ba^{131}, and these curves were used to assign a particular fissioning nucleus to these products at all energies. In all instances the measured ranges were first corrected for the difference between the actual and effective charges on the basis of reported charge dispersions.[1,8,10,27]

The resulting values of A_F for Sr^{83} and Ba^{131} are plotted as a function of proton energy in Fig. 8. The error bars are based solely on the uncertainties in the measured ranges. It is seen that the mass number of the average fissioning nucleus is constant at about 234 up to 1 GeV, decreases to approximately 170 at 4.5 GeV, and remains nearly constant thereafter. The corresponding Z value of this nuclide exhibits an accompanying decrease from 92 to approximately 72. The same value of A_F is consistent with the ranges of both Sr^{83} and Ba^{131} up to 4.5 GeV. At higher energies a smaller A_F appears to be required for Sr^{83}.

The apparent decrease in the mass of the fissioning nucleus by some 65 amu between 1 and 4.5 GeV implies a profound change in the nature of the proton-nucleus interaction. The probability of interactions involving excitation energies of some 200 MeV, which are the ones leading to these fissioning nuclei at the lower energies, would have to be sharply reduced and be accompanied by a large increase in the probability of interactions involving deposition energies in excess of 500 MeV. At the same time, however, the probability of transfers of about 100 MeV cannot be materially affected, in view of the nearly constant yields of the neutron-excessive fission products. Such a change in the deposition-energy spectrum appears to be rather unreasonable in the light of results at lower energies.[29]

The required E^* values of 500–600 MeV needed to form fissioning nuclei in the 160–170 mass range are not consistent with the values inferred from the recoil properties. Although the E^* values summarized in Table V do indeed increase in the required manner up to 2–3 GeV, their behavior at higher energies cannot be reconciled with that of the A_F values. A possible resolution of this discrepancy lies in the assumption that the $v_{\parallel}$-E^* relation embodied in Eq. (3) breaks down at high energies. We have no reason, however, to assume such a breakdown, and the E^* values derived from Sr^{91} and Ba^{140}, in fact, appear to be perfectly reasonable over the entire energy range up to 11.5 GeV.

Another argument against a fission mechanism can be made on the basis of cross-section considerations. The A_F values plotted in Fig. 8 indicate that the fission of tantalum should be quite similar to the alleged high-energy fission process

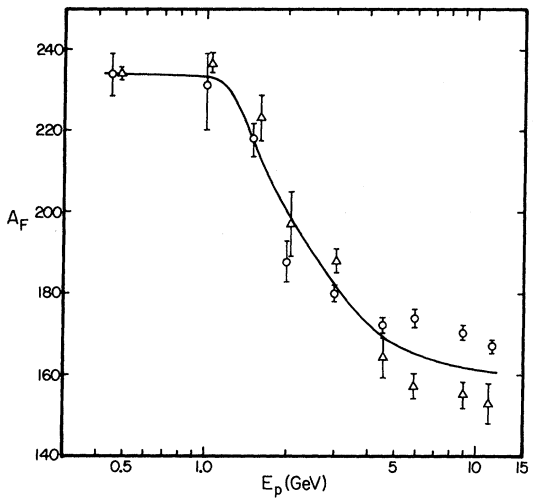

FIG. 8. Energy dependence of the mass number of the average fissioning nucleus leading to the formation of Sr^{83} (Δ) and Ba^{131} (o).

leading to the neutron-deficient products. The cross section for the formation of Ba^{131} from Ta has been measured with 340-MeV protons as 0.013 mb.[32] The shape of the mass-yield curve at this energy indicates that Ba^{131} is primarily a fission product and is, moreover, fairly close to being the most probable product at this mass number. Since the fission cross sections of various heavy elements increase by at most a factor of 2 between 0.34 and 6 GeV[33] and decrease at higher energies, the expected fission yield of Ba^{131} from Ta at 12 GeV is no more than 0.03 mb. This is a factor of nearly 300 smaller than the yield of Ba^{131} from uranium at this energy. If fission of a nucleus such as Hf^{164} were the main reaction mechanism, the probability of reaching such a product in the interaction of uranium with 12-GeV protons would have to exceed that of forming it from Ta by a comparable factor. Although cascade-evaporation calculations are not available for 12-GeV protons, the results at lower energies indicate that such a large ratio is rather unreasonable. It thus appears that binary fission cannot contribute in a substantial way to the yield of the neutron-deficient products at 12 GeV.

B. Spallation

A number of discrepancies between our results and those expected for spallation are qualitatively apparent. It is clear that in order to produce barium and strontium products by this mechanism very lengthy cascade-evaporation chains, involving high deposition energies, are required. We have estimated, on the basis of evaporation calculations described below, that E^* values of 900 and 1800 MeV are most effective in leading to products with $A = 131$ and 83, respectively. If spallation were of primary importance above 4 GeV, the E^* values derived from the recoil properties should reflect these large energy transfers. We have already dwelled on the fact that this is not the case and once again a breakdown of the $v_{\parallel}$-E^* relation would have to be assumed.

One of the basic features of high-energy interactions is that the average deposition energy increases rather slowly with bombarding energy. Monte Carlo cascade calculations[29] indicate that E^* for uranium increases by about 200 MeV per GeV for bombarding energies between 1 and 2 GeV. A deposition-energy difference of 900 MeV, corresponding to barium and strontium formation, would thus seem to require a concomitant bombarding-energy difference of 4–5 GeV. If spallation were responsible for the observed decrease in range, this effect should thus be noticeable at a much higher bombarding energy for Sr^{83} than for Ba^{131}. Our results indicate that this is not the case.

In order to obtain quantitative information on the magnitude of the ranges and forward-to-backward ratios expected for spallation we have performed a Monte Carlo evaporation calculation. We have used a code based on the formalism of Dostrovsky, Fraenkel, and Friedlander[34] as modified by Porile and Tanaka,[35] and Porile[36] to calculate recoil properties. In the absence of high-energy cascade calculations we have assumed a single residual nucleus, Pb^{203}, to replace the distribution expected from the cascade. Extrapolation of existing cascade calculations[29] indicated that this nuclide should be a reasonable choice for interactions involving very high excitation energies. Evaporation calculations performed for different initial excitation energies indicated that values of 900 and 1800 MeV resulted in the highest yield of products with $A = 131$ and 83, respectively. These values were accordingly adopted for all subsequent calculations. The initial velocity that Pb^{203} would have acquired as a result of the intranuclear cascade was treated as an adjustable parameter. Calculations were performed for various values of both the forward, $v_{\parallel}$, and transverse, $v_{\perp}$, components of the impact velocity.

The evaporation calculation considered the emission of eight particles: n, p, d, t, He^3, He^4, Li^7, and B^{11}. The statistical weights of Li^7 and B^{11} were increased by factors of 4 and 10, respectively, to simulate the emission of all fragments up to $A = 12$. The maximum probabilities of emitting these fragments in a single step of a typical evaporation sequence are approximately 1.2% and 0.2%, respectively. However, the effect of these

massive particles on the recoil ranges is magnified by the high momentum imparted by their evaporation.

At the end of the deexcitation process the ranges of all products with mass numbers between 81 and 85 or 128 and 133 were calculated from the recoil velocities. The range-energy relation due to Lindhard, Scharff, and Schiøtt[37] was used for products having kinetic energies below 10 MeV. Above this energy the relation given by Bridwell and Moak[25] was used. The range of each product was projected along the beam axis with the aid of the calculated recoil angle. At the end of 1000 iterations the results for all products lying in either of the above mass intervals were combined to improve the statistical accuracy and the values of FW and BW obtained by means of equations given elsewhere.[38]

The results of the calculation are summarized in Fig. 9, which shows the dependence of $2W(F+B)$ and F/B on $v_{\parallel}$ for different values of $v_{\perp}$. The trends displayed by these results are readily explainable. The F/B values increase with $v_{\parallel}$ as expected from Eq. (2). For a given $v_{\parallel}$, F/B varies inversely with $v_{\perp}$. This behavior is expected from

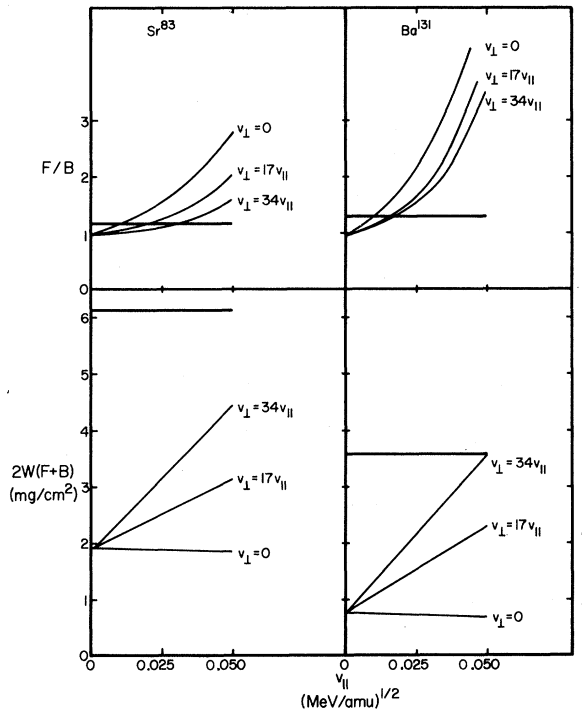

FIG. 9. Results of the spallation calculation of $2W(F+B)$ and F/B values of products with $A=81-85$ (left panel) and $A=128-133$ (right panel). The calculated values are plotted as a function of $v_{\parallel}$ for different values of $v_{\perp}/v_{\parallel}$. The experimental values for Sr^{83} and Ba^{131} are shown as the heavy horizontal lines.

a more complete form of Eq. (2), which includes a term in $\eta_{\perp}^2$ whenever the range-velocity exponent N is different from unity. It is also seen that the ranges increase with $v_{\perp}$. Once again, this follows from the fact that N is larger than unity in the energy region of interest.

The calculated ranges for $v_{\perp}=0$ are seen to be much smaller than the experimental values for both Sr^{83} and Ba^{131}. In an effort to determine whether it was at all possible to reproduce the experimental results the calculation was repeated for various values of $v_{\perp}$. It is known[28] that the ratio of $v_{\perp}/v_{\parallel}$ varies inversely with $v_{\parallel}$ and that values as large as 5 to 10 are possible for very small values of $v_{\parallel}$. Figure 9 indicates that the calculation does not yield the correct values of $2W(F+B)$ and F/B even for much larger values of this ratio. It should be pointed out that values of $v_{\parallel}$ and $v_{\perp}$ can be found that will fit either the ranges of the F/B values. However, it appears to be impossible to fit both quantities with the same impact-velocity components unless unreasonably large values of $v_{\perp}$ are assumed. Even then the fit is unsatisfactory from the viewpoint of the required value of $v_{\parallel}$ and its relation to the average deposition energy. For instance, the results for Ba^{131} could be reproduced with $v_{\parallel}=0.03$ $(\mathrm{MeV/amu})^{1/2}$ and $v_{\perp}/v_{\parallel}=40$. On the basis of Eq. (3) this value of $v_{\parallel}$ corresponds to $E^*=150$ MeV which is much lower than the required value of 900 MeV. We conclude that spallation cannot be primarily responsible for the formation of the neutron-deficient products at high energies.

C. Fragmentation

Since fragmentation is not as yet a very well characterized process, we shall begin by defining it in the context of the present experiment. We assume that this process involves the emission of a light fragment on a time scale comparable to that of the intranuclear cascade. This implies that there is a correlation between the direction of the emitted fragment and that of the propagation of the cascade so that the fragment is preferentially ejected into the forward hemisphere. This is precisely the situation for the emission of Na^{24} from the interaction of Bi with 2.9-GeV protons as determined from differential-range and angular-distribution measurements by Cumming *et al.*[39] A cascade that involves the emission of a fragment may be expected to form a highly excited residual nucleus. This nucleus would then deexcite to a final product by a long evaporation chain. This product would tend to lie on the neutron-deficient side of stability because spallation preferentially leads to this type of nuclide. Conservation of mo-

mentum dictates that the heavy product have a lower F/B value than it would in the absence of the preferential forward emission of a fragment. Finally, the recoil range of the heavy product should be larger then expected for spallation provided products comparable in mass to prompt fragments are evaporated with much smaller probability than that for the occurrence of fragmentation. On the other hand, the range should be smaller than expected for binary fission both because of the relatively low mass of the emitted fragment and the fact that it is emitted with an energy only half as large as the classical Coulomb barrier.[39,40] This description of fragmentation closely resembles that previously given by Crespo, Alexander, and Hyde[40] and Alexander, Baltzinger, and Gadzik,[2] and is consistent with the original formulation by Wolfgang *et al.*[41]

The present results are qualitatively consistent with fragmentation as defined above. The drop in the Ba^{131} and Sr^{83} range curves occurs at approximately the same energy as the peak in their forward-to-backward ratios. Fragmentation necessarily demands this behavior, since both effects result from the forward ejection of a light fragment. The concomitant decrease of the E^* values is readily explainable, since it follows from the corresponding decrease in $v_{\parallel}$, which in turn arises from the forward emission of the fragment. The anomalous behavior of the deposition energies thus follows naturally from the assumed mechanism and does not require the somewhat artificial assumption of a breakdown of the $v_{\parallel}$-E^* relation. The magnitudes of the ranges above 6 GeV are seen to be larger than predicted by spallation and smaller than expected for fission, in accord with the above-mentioned expectation.

We have made a more quantitative comparison of our results with those expected for fragmentation by means of a modified form of the evaporation calculation described in the last section. In addition to the cascade and evaporation steps, each interaction was assumed to involve the emission of a Na^{24} fragment having properties based on the report by Cumming *et al.*[39] The velocity components of the residual nucleus resulting from an intranuclear cascade accompanied by fragment emission were determined from the energy and angle of the emitted Na^{24} as well as from the values assumed for the nucleon-nucleon cascade. The Monte Carlo evaporation calculation was then performed in the manner described above using Pb^{203} as the starting nucleus, excited to the same energies as before. Our procedure thus allowed for the emission of 12 cascade nucleons, in addition to that of Na^{24}.

The assumed properties of the Na^{24} fragment are summarized in Table VI. Since Cumming *et al.*[39] measured differential ranges at three angles, it was assumed for simplicity that these were the only angles at which the fragment could be emitted. The fractional emission probabilities per unit solid angle at 15, 90, and 165° to the beam were obtained from the measured angular distribution. The differential ranges at each of these angles have been analyzed[39] in terms of Gaussian velocity distributions, and the mean velocities and their standard deviations are summarized in Table VI. The listed properties of Na^{24} were incorporated in the Monte Carlo calculation, and appropriately weighted random numbers were used to select a particular angle and velocity of Na^{24} for each iteration. The corresponding velocity components of the Pb^{203} residual nucleus were then obtained by conservation of momentum. The resulting values were added to those from the evaporation step as well as to those assumed for the cascade step to obtain the final product velocities. The ranges were obtained in the manner outlined above and the results are again based on 1000 iterations. As before, the calculation was repeated for several values of $v_{\parallel}$ and $v_{\perp}$.

The results of this calculation are summarized in Fig. 10. The dependence of $2W(F+B)$ and F/B on $v_{\parallel}$ and $v_{\perp}$ is qualitatively similar to that shown in Fig. 9 for spallation. One noticeable difference is that the F/B values are less than unity when $v_{\parallel}=0$. This is the expected consequence of the preferential forward emission of a light fragment. In addition, the ranges are 50–100% larger than those calculated for spallation, reflecting the emission of a massive fragment.

The results presented in Fig. 10 indicate that

TABLE VI. Assumed properties of the Na^{24} fragmentation product (Ref. 39).

Laboratory angle (deg)	Fractional emission probability (per unit solid angle)	Mean velocity $(\text{MeV/amu})^{1/2}$	Standard deviation $(\text{MeV/amu})^{1/2}$
15	0.45	2.17	0.39
90	0.34	1.98	0.37
165	0.21	1.78	0.33

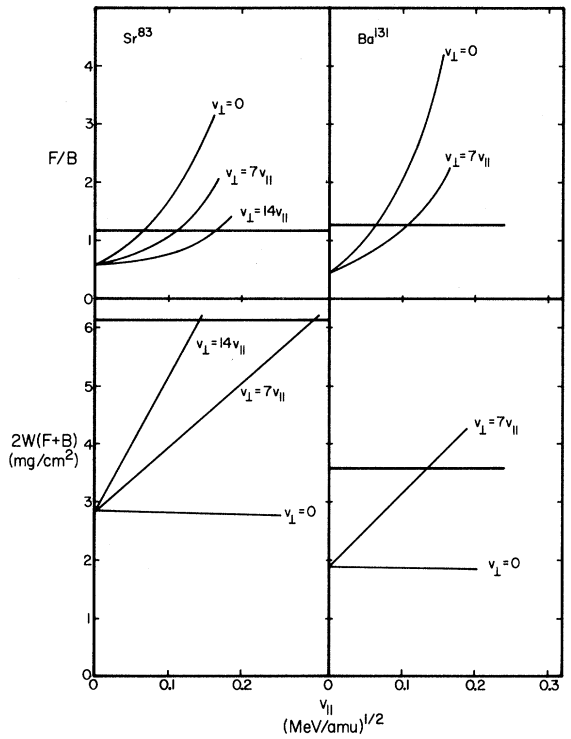

FIG. 10. Fragmentation calculation of ranges and forward-to-backward ratios. See Fig. 9 for details.

although the inclusion of Na^{24} emission leads to considerable improvement between the calculated and experimental ranges, the former are still low by nearly a factor of 2 when $v_\perp = 0$. The inclusion of $v_\perp$ clearly improves the agreement. The calculated values of both $2W(F+B)$ and F/B for $A \sim 131$ are in good agreement with the corresponding results for Ba^{131} for $v_\parallel = 0.12$ (MeV/amu)$^{1/2}$ and $v_\perp = 7v_\parallel$. Let us determine if these particular velocity values are physically reasonable.

The value of $v_\parallel$ corresponds to an average deposition energy of 600 MeV. This is in fair agreement with the 900-MeV value suggested by the evaporation calculation, indicating that $v_\parallel$ is of the correct magnitude.

The required value of $v_\perp$ is rather large although not outside the range predicted by Monte Carlo calculations[28,29] for interactions involving low momentum transfers. Let us compare this value of $v_\perp$, 0.84 (MeV/amu)$^{1/2}$, with some of the other average velocities involved in the interaction. The calculated value of V, the average velocity of the product due to evaporation, is 0.25 (MeV/amu)$^{1/2}$. The velocity the residual nucleus acquires as a result of Na^{24} emission is 0.24 (MeV/amu)$^{1/2}$. It is thus apparent that $v_\perp$ is the dominant velocity vector. One would therefore expect the angular distribution of such a fragmentation product to feature a strong peak at sideward angles, a result that has not been observed at a bombarding energy of 2.2 GeV.[14,42] For instance, the angular distribution and differential range of Ba^{131} from the interaction of uranium with 2.2-GeV protons[14] indicates that $v_\perp/v_\parallel \sim 1.5$. Although fragmentation is not the dominant mechanism at this low an energy our results suggest that it already is an important contributor to the Ba^{131} yield so that there should be some experimental indication of large $v_\perp$ values.

The calculation for $A \sim 83$ is in even more severe disagreement with experiment. In order to match the results obtained for Sr^{83} it is required that $v_\parallel = 0.15$ (MeV/amu)$^{1/2}$ and that $v_\perp/v_\parallel \sim 14$. Such a large velocity ratio is physically unreasonable and furthermore does not agree with the expected[28] inverse dependence of $v_\perp/v_\parallel$ on $v_\parallel$. Furthermore, the required value of $v_\parallel$ corresponds to $E^* = 750$ MeV, which is substantially lower than 1800 MeV, the value predicted by the evaporation calculation.

It is thus apparent that although the inclusion of fragment emission in the cascade-evaporation formalism vastly improves the agreement with experiment, serious discrepancies still remain. This is really not very surprising, since our model for fragmentation is admittedly crude. There is obviously no reason to assume that Na^{24} is the only possible emitted fragment. The recent report by Poskanzer, Butler, and Hyde,[43] in fact, indicates that the preferential forward emission of light fragments is a fairly common occurrence. A more complete calculation would be valuable but not before cascade calculations at 6–12 GeV have been performed.

If fragmentation is indeed the principal mechanism for the production of neutron-deficient products lying in the "fission" region, there must be a correspondence between their formation cross sections and those of the emitted light fragments. The excitation functions shown in Fig. 5 suggest an average fragmentation cross section of about 7 mb per mass number over the mass range 83 to 131. These masses undoubtedly do not define the limits of the mass region where fragmentation may be expected to contribute. The decrease in the range of neutron-deficient products at high energies has also been detected for lighter elements such as bromine[4] and undoubtedly also occurs above $A = 131$. For the purposes of the present estimate we assume a fragmentation contribution of 7 mb/mass number extending from $A = 70$ to 150, resulting in a total cross section of 560 mb. This estimate compares favorably with that of 600 mb obtained by subtracting the total fission cross section measured with mica track detectors at 29 GeV[33] from the corresponding mass-yield curve[10] integrated

from $A = 70$ to 150. The light fragments whose emission leads to the above products can be expected to range from about $A = 20$ to 60. The results of the fragmentation calculation indicate that lighter fragments are not massive enough to account for the observed ranges of the heavy residues unless more than one fragment is emitted per interaction. The upper limit of this mass range is somewhat arbitrarily set just slightly below the lower limit of that of the heavy residual nuclei. A recent survey[10] of the high-energy fission of uranium indicates an integrated cross section of approximately 530 mb over the above mass range at 10–30 GeV. The cross sections of light fragments and heavy neutron-deficient products are thus consistent with fragmentation, provided the assumption is made that virtually all products with $A = 20$–60 are the result of this process.

V. CONCLUSIONS

The energy dependence of the recoil properties of a number of barium and strontium nuclides formed in the interaction of uranium with high-energy protons has been measured between 0.45 and 11.5 GeV. The ranges of the neutron-deficient products decrease by about a factor of 2 between 1 and 5 GeV and exhibit only a minor energy dependence outside this interval. The forward-to-backward ratios increase up to 2–3 GeV and decrease thereafter.

The properties of the neutron-deficient products are consistent with a binary-fission process up to 1 GeV at which point a contribution from another type of process becomes noticeable. This process becomes dominant by 4 or 5 GeV. We have examined in detail the possibility that fission, spallation, or fragmentation could account for the results. Of these three mechanisms only fragmentation can explain all the data, although some discrepancies remain with our simple calculation.

Considerations based on yield measurements indicate that the total fragmentation cross section of uranium (per binary event) at 10–30 GeV is 500–600 mb. This process thus accounts for a substantial fraction of the total reaction cross section and should be detectable in a counter experiment. The experiments by Remsberg *et al.*,[44] in which coincident fragments from the fission of U^{238} by 2.9-GeV protons were measured with solid-state detectors, showed no evidence for fragmentation. This may have been due to a combination of experimental restrictions coupled with too low a bombarding energy. If our explanation of the observed phenomena is correct, then a counter experiment performed at higher energies should give evidence for forwardly emitted light fragments in coincidence with heavy fragments of rather low energy. Although we feel that fragmentation is the main process leading to the formation of neutron-deficient products above 5 GeV, spallation and fission undoubtedly do contribute to some extent. By the same token, fragmentation probably also contributes in a minor way to the yield of these products below 1 GeV. Our discussion should be viewed as applying to the main but not the entire features of the reaction mechanism.

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Application of the Statistical Theory to Heavy-Ion Reactions

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The cross section of heavy-ion reactions is studied in the framework of the statistical theory of nuclear reactions. An expression is obtained for the probability of production of different isotopes in heavy-ion collisions. A comparison with the experimental data obtained recently in Dubna is given.

I. INTRODUCTION

Heavy-ion reactions in which several nucleons are transferred from target to projectile, or vice versa, are the subject of many recent investigations.[1-3] The mechanism of these reactions is usually classified in terms of the distance of closest approach of the colliding nuclei. When the bombarding energy is well above the Coulomb barrier there is a considerable probability for an intermediate system to be formed. According to Newton,[3] the relative probability of the direct and resonant mechanisms are such that the cross section of compound-nucleus formation is roughly about 500 mb, while that for single-nucleon transfer is about 5 mb.

Recently, Oganesyan *et al.*[4] investigated the rate of production of various isotopes in heavy-ion reactions above the Coulomb barrier. These authors found that the reaction cross section corresponding