Formation of isobaric nuclides with $A = 131$ in the interaction of ^{238}U with 0.8–11.5 GeV protons

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Cross sections and thick-target recoil properties of ^{131}Ce, ^{131}La, ^{131}Ba, and several neighboring Ba nuclides formed in the interaction of ^{238}U with 0.8–11.5 GeV protons have been determined. The results reveal a dichotomy between ^{131}Ba and the more neutron-rich products on the one hand, and the more neutron-deficient ones on the other. The latter have rising excitation functions, forward-to-backward ratios (F/B) that peak at 3 GeV, and ranges that decrease abruptly just below this energy. The former have decreasing excitation functions as well as F/B and ranges that decrease slowly and featurelessly with increasing proton energy. The energy dependence of F/B as well as the changes in the angular distributions of the very neutron deficient products observed between 3 and 11.5 GeV are explained in terms of a change in the nature of near-central proton-nucleus interactions. At the lower energies the interaction consists of a series of nucleon-nucleon collisions while at high energies the proton interacts collectively with all the nucleons lying in its path. The dropoff in the mean ranges of these same nuclides and the accompanying broadening of the spectra indicate a transition between fission and deep spallation. The possible connection between these two effects is discussed. Several experiments to test the proposed model are suggested.

[NUCLEAR REACTIONS Measured σ, range, and F/B of ^{131}Ce, ^{131}La, ^{131}Ba, ^{128}Ba, $^{133}Ba^m$, $^{135}Ba^m$, and ^{140}Ba formed in interaction of ^{238}U with 0.8–11.5 GeV protons. Proposed explanation for change in properties of very neutron deficient products.]

I. INTRODUCTION

In a previous publication from this laboratory[1] we reported the discovery of a transition in the properties of neutron-deficient products with $A \sim 80-140$ formed in the interaction of ^{238}U with high-energy protons. This transition occurred at approximately 3 GeV and manifested itself in a peak in the ratio of forward-to-backward emission (F/B) and in a sharp decrease in recoil range. These phenomena were interpreted as resulting from a change in the reaction mechanism from binary fission to deep spallation, a process involving the formation of products far from the target nuclide as a result of the emission of nucleons, light aggregates, and fragments. By contrast, the ranges and F/B ratios of neutron-excess products in this mass region showed little, if any, variation with energy between 1 and 11.5 GeV, indicating that these nuclides were formed in a fission process over the entire energy range. Earlier work[2–6] had already shown that the ranges of neutron-deficient products were distinctly shorter at 6 GeV and above than below 1 GeV. Other experiments[7–15] had similarly shown that the ranges of neutron-deficient products were substantially shorter than those of neutron-excess nuclides of comparable atomic number at multi-GeV energies. More recently, evidence for a similar transition at ~3 GeV has been observed for various light fragments formed in the interaction of ^{238}U with high-energy protons[16,17] as well as for a variety of products from gold.[18] Recent angular distribution measurements[19–21] indicate that the decrease in F/B observed above 3 GeV appears to be associated with a change in the angular distributions from forward peaked to sideward peaked.

The results of Bég and Porile[1] were obtained for cumulatively formed products and so reflect the average behavior of a given product and its isobaric progenitors. For instance, the results reported for ^{131}Ba actually represent the average recoil properties of ^{131}Ba, ^{131}La, and ^{131}Ce, weighted by the respective formation cross sections. These results do not provide enough information to determine whether the transition occurs at the same energy for all isobaric nuclides or whether it occurs at different energies depending on the composition of the product. If the latter alternative holds, the width of the peak in F/B will be narrower and the dropoff in range sharper than was observed for cumulative products.[1] Since the nature of the observed transition is not as yet fully understood, it is of some importance to characterize it as completely as possible. Furthermore, although it is fairly well established that the transition between long and short range products occurs between one and two Z units on the neutron-deficient side of stability for bombarding energies of 10 GeV or higher,[10,11,13] it is not known whether the peak in F/B is also first seen for products of this same composition. If the decrease in range and the peak in F/B are to be ascribed to the same phenomenon, there should be a close correspondence between the products exhibiting these two effects. With the exception of the lightest fragments,[16] whose F/B ratios peak but whose ranges do not drop off near 3 GeV, there does indeed appear to be such a correspondence. However, the experimental results available to date are too sparse to permit any definitive conclusions to be drawn. Results for isobaric neutron-deficient nuclides would be helpful in filling the existing gaps. With these ends in mind, we have examined the energy dependence of the thick-target recoil properties and cross sections for the formation of ^{131}Ba, ^{131}La, ^{131}Ce, and several neighboring Ba nuclides in the interact on of ^{238}U with 0.8–11.5 GeV protons.

II. EXPERIMENTAL

The experimental procedures were dictated by the half-lives of the isobaric nuclides of interest. The following is the genetic relationship of these nuclides:

$$^{131}\mathrm{Ce}\xrightarrow[t_{1/2}=10\,\mathrm{min}]{}{}^{131}\mathrm{La}\xrightarrow[t_{1/2}=59\,\mathrm{min}]{}{}^{131}\mathrm{Ba}\xrightarrow[t_{1/2}=11.7\,\mathrm{d}]{}.$$

Two sets of experiments were performed, one for the determination of ^{131}Ce, and the other for that of ^{131}La, ^{131}Ba, and the other Ba nuclides.

Most of the irradiations were performed with 1.0–11.5 GeV protons in the circulating beam of the Zero Gradient Synchrotron (ZGS) at Argonne National Laboratory. In addition, a few irradiations with 0.8-GeV protons were performed in the nuclear chemistry irradiation facility in area B of the Los Alamos Meson Physics Facility (LAMPF). Target stacks consisted of 20 μm thick depleted uranium foil sandwiched between five 20 μm thick aluminum foils of high purity (99.999%), three on the upstream side, and the remaining two on the downstream side. The inner pair served as recoil catchers, the outer pair as guards, and the middle foil on the upstream side as the beam intensity monitor. All foils were cut to the same area and carefully aligned to ensure that they intercepted the same number of protons. The irradiations designed to yield results for ^{131}Ce had a duration of 5–15 min and 24 separate experiments were performed while those designed for La/Ba were 18 in number, and ranged in duration from 5 to 65 min.

Following irradiation the target and catcher foils were separately dissolved in acid and a radiochemical separation appropriate to each type of experiment was performed.[11] In the cerium experiments, the element was rapidly separated as $Ce(IO_3)_4$. The chemical yield was determined and the samples were set aside for a sufficiently long time (≥ 7 h) to ensure virtually complete decay of ^{131}Ce to ^{131}Ba. At this point the samples were dissolved and Ba was separated and purified. In the La/Ba experiments Ba was quickly separated. Lanthanum was allowed to remain in solution long enough to permit essentially complete decay of ^{131}La at which time Ba was separated.

The disintegration rates of the various nuclides of interest were determined by γ-ray spectrometry with a calibrated Ge(Li) detector operated in conjunction with a 4096 channel analyzer. The decay properties of the nuclides of interest[22–27] are summarized in Table I. As implied above, ^{131}Ce and ^{131}La were assayed via their ^{131}Ba decay product. The spectra were analyzed with the code SAMPO[28] in order to obtain the disintegration rates and the latter were extrapolated to the end of bombardment on the basis of the tabulated half-lives. The results were corrected for reduction in photopeak intensity due to coincidences between the detected and other γ rays emitted by a given nuclide on the basis of the formulation of McCallum and Coote.[29]

The beam intensity was determined by means of the ^{27}Al(p, $3pn$) monitor reaction.[30] The assay

TABLE I. Decay properties of observed nuclides.

Nuclide	Half-life	γ ray (keV)	Branching ratio, %
^{128}Ba	2.43 d	443.0 [a]	35
		273.1	21 [b]
^{131}Ba	11.7 d	123.7	28.2 [c]
		496.2	46.5 [d]
^{133}Ba^m	39.9 h	276.1	17.0
^{135}Ba^m	28.7 h	268.2	15.6 [e]
^{140}Ba	12.8 d	537.3	20

[a] This γ ray is that of daughter (^{128}Cs) in equilibrium with ^{128}Ba.

[b] In view of the discrepant reports about the branching ratio of this γ ray it was only used for recoil property determinations. The 443.0-keV γ ray was used for cross section determinations.

[c] This branching ratio was obtained from the experimental relative intensities of the 123.7- and 496.2-keV γ ray to be 46.5%. Both 123.7- and 496.2-keV γ rays were used for cross section and recoil property determinations.

[d] This branching ratio is based on the total transition intensity reported in Ref. 23 and the conversion coefficient from Ref. 24.

[e] Branching ratio computed on basis of the conversion coefficient reported in Ref. 25.

of the aluminum foils and the analysis of the data were based on the procedures described above.

III. RESULTS

The cross sections of the $A = 131$ products were derived from the data on the basis of equations[31] appropriate to the genetic relationship between these nuclides. Corrections were applied for growth and decay of progenitors during irradiation and up to the time of parent-daughter separation. The results obtained for ^{131}Ce were corrected for variations in beam intensity. This correction could be neglected for the longer-lived products. The possible formation of the 5 min isomer of ^{131}Ce was not incorportated in the analysis. It has been shown on the basis of more complete measurements[11] that this leads to an ~10% uncertainty in the ^{131}Ce and ^{131}La cross sections. The other Ba nuclides for which results are reported had no special problems associated with progenitor decay and the results were obtained by means of standard relationships. The cross sections were corrected for the contribution of secondary reactions on the basis of the data reported by Yu and Porile.[11] The correction amounted to ~15% for ^{140}Ba and to ~5% for the neutron-deficient products, for which it actually reflects the secondary effect on the monitor reaction.

The quantities determined in recoil experiments of this type are the fraction of the total disintegration rate of a given nuclide observed in the forward and backward catchers, denoted by F and B, respectively. The recoil properties of interest are the experimental range, $2W(F+B)$, where W is the target thickness, and the ratio of forward-to-backward emission, F/B. The ranges were reduced by 3% to correct for scattering at the interface between target and catcher.[32]

Two to five replicate experiments were performed at each energy. An examination of replicate results showed that in a number of instances there appeared to be systematic differences between different experiments. For instance, the ranges of all nuclides determined in a particular experiment might be uniformly higher than those obtained in another one. A likely cause of such differences is nonuniformity in target thickness. Usually, there is not much that can be done about such effects except to increase the number of replicate determinations. However, in the present study it proved possible to develop a normalization procedure for the La/Ba runs that virtually eliminated systematic differences between replicate experiments and substantially reduced the overall uncertainties. This procedure takes advantage of the fact that the cross sections, ranges, and F/B ratios of ^{140}Ba, a typical low-deposition energy fission product, are virtually independent of bombarding energy in the GeV regime. A linear least-squares fit to the data for this nuclide permitted a determination of the deviations of individual results from the expected trend. Moreover, the availability of ^{140}Ba data from other comparable experiments[1,15] improved the statistical accuracy of the fit.

Figure 1 shows the data points for ^{140}Ba and the least-squares fits to these values. In obtaining these fits we did not include points that differed by more than 3 σ from the lines. The fit to the cross sections was not extended below 2 GeV because the excitation function turns up at lower energies. While the points exhibit a fair amount of scatter, they nonetheless show that the energy dependence of the plotted quantities is indeed small. The fitted lines were used to correct the data for ^{131}La and the Ba isotopes other than ^{140}Ba by multiplying the result obtained in a given experiment by the corresponding ratio of fitted to experimental values for ^{140}Ba. This procedure substantially improved the agreement between replicate determinations. For instance, the mean range of ^{128}Ba at 3 GeV changed from 4.33 ± 0.23 mg/cm^2 to 4.42 ± 0.08 mg/cm^2.

The weighted average values of the cross sections, recoil ranges, and F/B ratios are tabulated in Tables II–IV, respectively. The designations C and I indicate whether the products in question represent cumulative or independent yields, respectively. The contribution of ^{131}Ce to the ^{131}La activity proved too large to permit a meaningful determination of independent ^{131}La. Accordingly, only results for the cumulative formation of this nuclide are presented. The listed uncertainties are the larger of the standard deviations from the means of replicate determinations and estimates of the uncertainties in individual determinations. The latter are based on the uncertainties in the γ-ray counting rates as obtained from SAMPO, on those in chemical yield determination (3–5%), and for the cross sections, on those in detector efficiency (5%). In addition, a 5% error in target thickness uniformity was incorporated in the range and cross-section results obtained for ^{131}Ce and ^{140}Ba. For the other nuclides, this error was based on the normalization procedure described above.

Some of the present results may be compared with previous determinations. Friedlander *et al.*[33] determined the cross sections of some of the same products up to 3 GeV. Their results for ^{131}Ba (I) and ^{140}Ba (C) agree, on the average, to within 10% with the present data. However, their ^{131}La (C) cross sections are approximately 35% larger than

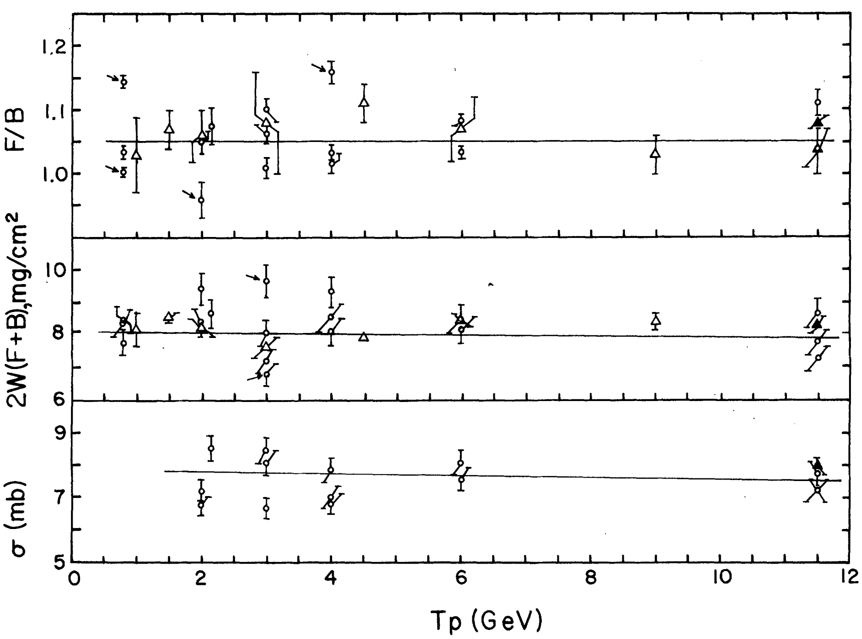

FIG. 1. Linear least-squares fit to ^{140}Ba data. From top to bottom: F/B, range, cross section; ○, present results; △, data from Ref. 1; ▲, data from Ref. 15; (⸸), points differing by more than 3σ from fitted lines, not included in fit.

those reported here. The results are generally in very good agreement with previous reports from our laboratory.[1,11,15] The only exception occurs for ^{128}Ba (*C*) whose cross sections were previously[1] reported to be some 70% larger than the present values. While the assay of this nuclide in the earlier work was based on a different technique, namely determination of the positrons emitted by the ^{128}Cs daughter via coincidence counting of the annihilation quanta, we are at a loss to explain such a large discrepancy. Fortunately, this difference is of no consequence to the central aspects of the present work.

IV. DISCUSSION

A. Energy dependence of measured and derived quantities

The energy dependence of the cross sections, F/B ratios, and ranges is displayed in Fig. 2. The products are arranged, from bottom to top, in order of increasing neutron deficiency, as given by the value of $(Z_A - Z_{\rm eff})$. The quantity $Z_{\rm eff}$ is the average *Z* value of all nuclides contributing to a cumulatively formed product weighted by their respective cross sections. In the case of independent products, $Z_{\rm eff}$ is just the atomic number of

TABLE II. Cross sections (mb) for formation of products in the interaction of ^{238}U with protons.

T_p (GeV)	^{131}Ce (*C*) [a]	^{131}La (*C*)	^{131}Ba (*I*) [a]	^{128}Ba (*C*)	^{133}Ba^m (*I*)	^{135}Ba^m (*I*)	^{140}Ba (*C*)
0.8	0.20 ± 0.04	1.58 ± 0.09	4.03 ± 0.20	1.04 ± 0.07	4.07 ± 0.27	4.12 ± 0.29	11.37 ± 0.64
1.0	0.88 ± 0.04						
2.0	1.56 ± 0.08	3.05 ± 0.08	2.44 ± 0.09	2.00 ± 0.10	2.50 ± 0.11	2.27 ± 0.10	8.65 ± 0.61
3.0	3.35 ± 0.16	4.75 ± 0.32	2.13 ± 0.10	2.84 ± 0.08	2.07 ± 0.06	2.02 ± 0.06	8.88 ± 0.68
4.0	4.01 ± 0.34	6.00 ± 0.79	2.50 ± 0.13	3.63 ± 0.19	1.80 ± 0.05	1.80 ± 0.05	8.59 ± 0.30
6.0	3.58 ± 0.36	7.06 ± 0.35	2.62 ± 0.32	3.89 ± 0.16	1.56 ± 0.06	1.58 ± 0.06	9.22 ± 0.31
11.5	3.61 ± 0.15 [b]	6.52 ± 0.25 [b]	2.43 ± 0.26 [b]	3.75 ± 0.17	1.53 ± 0.07	1.55 ± 0.07	8.70 ± 0.24

[a] The symbols (*C*) and (*I*) stand for cumulative and independent yields, respectively.
[b] Data from Ref. 11 are averaged with the present data after adjustment for differences in assumed branching ratios.

TABLE III. Experimental recoil ranges $[2W(F+B)]$ of observed nuclides in mg/cm^2 of uranium.

T_p (GeV)	^{131}Ce	^{131}La	^{131}Ba	^{128}Ba	^{133}Ba^m	^{135}Ba^m	^{140}Ba
0.8		7.05 ± 0.54	6.16 ± 0.22	6.17 ± 0.28	7.23 ± 0.10	7.25 ± 0.10	7.93 ± 0.23
1.0	8.12 ± 0.37						
2.0	7.74 ± 0.49	4.68 ± 0.24	7.07 ± 0.15	5.64 ± 0.10	7.29 ± 0.10	7.46 ± 0.10	8.51 ± 0.30
3.0	3.99 ± 0.16	3.71 ± 0.57	6.38 ± 0.26	4.42 ± 0.08	6.99 ± 0.11	7.19 ± 0.10	7.40 ± 0.55
4.0	3.85 ± 0.11	2.99 ± 0.06	6.30 ± 0.15	3.90 ± 0.08	6.93 ± 0.10	7.20 ± 0.10	8.25 ± 0.25 [a]
6.0	3.11 ± 0.08	2.70 ± 0.07	6.09 ± 0.24	3.51 ± 0.08	6.85 ± 0.10	7.24 ± 0.10	8.05 ± 0.29
11.5	2.55 ± 0.16 [a]	3.05 ± 0.21 [a]	5.28 ± 0.20 [a]	3.20 ± 0.08	6.69 ± 0.14	7.03 ± 0.15	7.77 ± 0.27

[a] Data from Ref. 11 are averaged with the present data.

the nuclide in question. The most stable charge at mass number A is designated Z_A. The numerical values of $(Z_A - Z_{\text{eff}})$ were taken from the work of Yu and Porile[11] and are valid at 11.5 GeV.

Figure 2 gives a striking indication that the products of interest may be divided into two distinct groups on the basis of all the plotted quantities. Nuclides with $(Z_A - Z_{\text{eff}}) \geq -1.7$ are thus characterized by falling excitation functions and decreasing but featureless F/B and range curves. On the other hand, nuclides with $(Z_A - Z_{\text{eff}}) \leq -3.6$ have rising excitation functions, F/B, that peak in the vicinity of 3 GeV, and ranges that drop sharply just below this energy. Another indication of this dichotomy is shown in Fig. 3. Plotted here are ratios of cross sections, ranges, and F/B at appropriate energies as a function of distance from stability. A clear difference between the most neutron-deficient nuclides and the rest is once again apparent. The cross sections of ^{131}Ba–^{140}Ba are thus about a factor of 2 lower at 6–12 GeV than at 0.8 GeV, while those of the most neutron-deficient products are much higher at the higher energies. The ranges of ^{131}Ba–^{140}Ba are only 5–15% lower at 6–12 GeV than at 0.8–2 GeV. On the other hand, the ranges of the most neutron-deficient products are a factor of 2 lower at the higher energies. Finally, the F/B of ^{131}Ba–^{140}Ba are practically equal at 11.5 and 3 GeV while those of ^{131}Ce, ^{131}La, and ^{128}Ba drop by 30% between these energies.

A detailed examination of the data indicates that the peak in F/B occurs at essentially the same proton energy in all the cases in which it is observed. The widths of the peaks are also closely comparable. It thus appears that the composition of a product determines whether F/B peaks or not, but if it does, the location of the peak is independent of composition. The rather striking correlation between the occurrence of this peak and the shape of the excitation function is novel but, in retrospect, perhaps not unexpected. It was pointed out long ago[34] that the shape of the excitation function depends on the deposition energy required to form the product in question. A rising excitation function thus requires a substantially higher energy transfer to the struck nucleus than a falling curve. The peak in F/B thus appears to occur only for products that are the result of highly inelastic interactions.

While the behavior of the F/B ratios is very uniform, that of the ranges is rather less so. Figure 2 indicates that the energy at which the steepest decrease in range occurs as well as the magnitude of the dropoff are somewhat variable. In addition, the ranges of the more neutron-deficient products that do not have a peak in F/B, ^{131}Ba in particular, decrease continuously over all or most of the energy interval. The variation of the ranges thus is more gradual than that of the other

TABLE IV. Energy dependence of F/B ratios.

T_p (GeV)	^{131}Ce	^{131}La	^{131}Ba	^{128}Ba	^{133}Ba^m	^{135}Ba^m	^{140}Ba
0.8		1.53 ± 0.09	1.35 ± 0.08	1.50 ± 0.02	1.33 ± 0.02	1.25 ± 0.02	1.05 ± 0.04
1.0	1.46 ± 0.08						
2.0	1.91 ± 0.07	1.85 ± 0.05	1.43 ± 0.03	1.72 ± 0.02	1.33 ± 0.05	1.24 ± 0.03	1.03 ± 0.03
3.0	1.92 ± 0.09	1.96 ± 0.10	1.34 ± 0.08	1.79 ± 0.02	1.25 ± 0.02	1.18 ± 0.02	1.05 ± 0.02
4.0	1.81 ± 0.09	1.88 ± 0.07	1.36 ± 0.04	1.75 ± 0.02	1.24 ± 0.03	1.17 ± 0.02	1.05 ± 0.03
6.0	1.58 ± 0.08	1.50 ± 0.10	1.35 ± 0.15	1.47 ± 0.02	1.18 ± 0.02	1.13 ± 0.02	1.06 ± 0.02
11.5	1.34 ± 0.04 [a]	1.27 ± 0.06 [a]	1.27 ± 0.08 [a]	1.31 ± 0.02	1.19 ± 0.03	1.14 ± 0.02	1.06 ± 0.03

[a] Data from Ref. 11 are averaged with the present data.

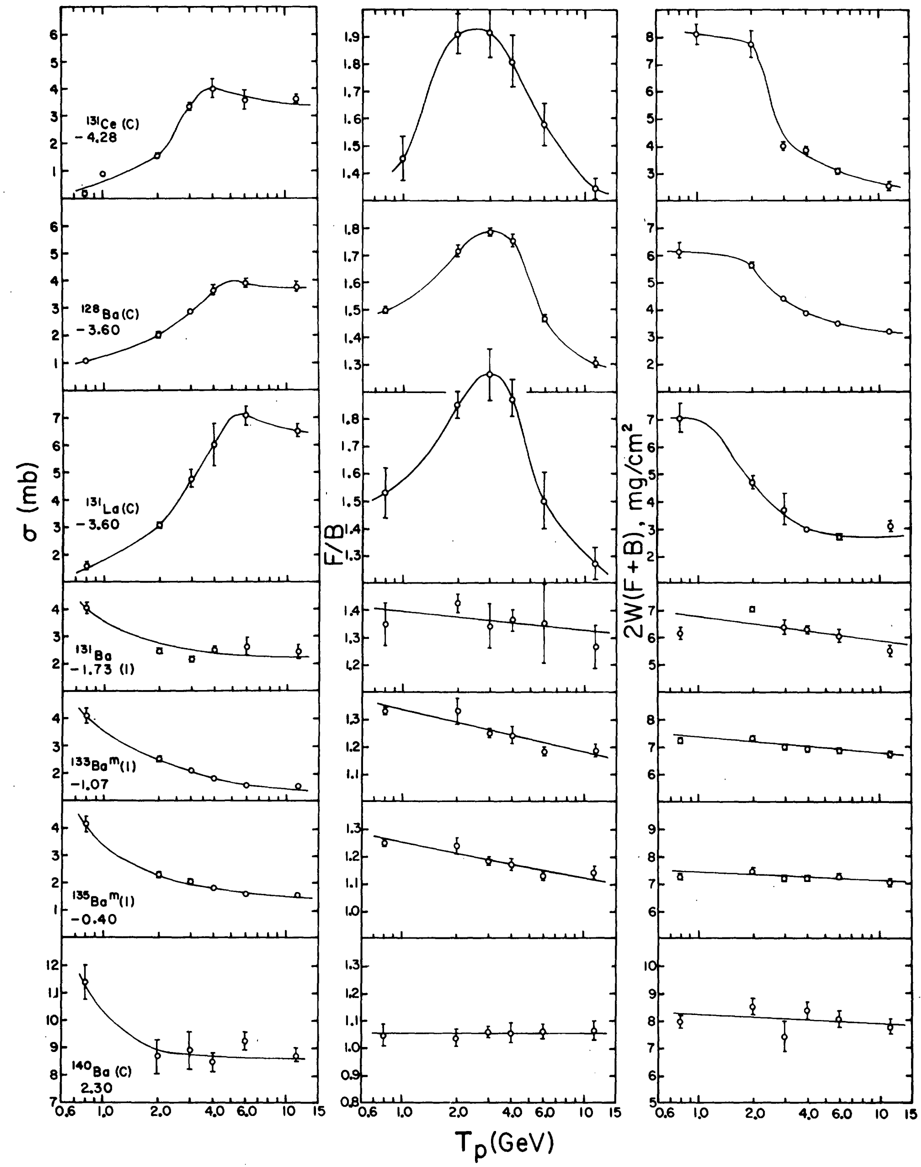

FIG. 2. Energy dependence of σ (left), F/B (middle), and range (right) of products from the interaction of ^{238}U with high-energy protons. The numbers listed in the left panels are the values of $(Z_A - Z_{eff})$ defined in text. The curves show the trends in the data.

quantities. This fact is also evident in Fig. 3, where it may be noted that the range ratio curve behaves in a more continuous manner than the curves for the F/B or cross-section ratios.

The measured recoil properties may be used to obtain the values of the mean forward component of velocity imparted to the struck nuclei in the initial interaction, $\langle v_{\parallel} \rangle$, by means of equations based on the two-step model of high-energy reactions.[7,17] The results have not been corrected for the possible overlap between the impact and breakup velocities. This effect is not expected to exceed 20% and does not qualitatively change the results.[35] Figure 4 displays the energy dependence of the $\langle v_{\parallel} \rangle$ values of the nuclei leading to the formation of the products of interest. As in the case of the other quantities determined in this experiment, the behavior of $\langle v_{\parallel} \rangle$ is qualitatively different for products with $(Z_A - Z_{eff}) \leq -3.6$ than for those with $(Z_A - Z_{eff}) \geq -1.7$. The nuclei giving rise to the former thus have $\langle v_{\parallel} \rangle$ that peak in the vicinity of 2 GeV while those leading to the formation of the latter decrease slowly and monotonically with increasing energy. An examination of the variation

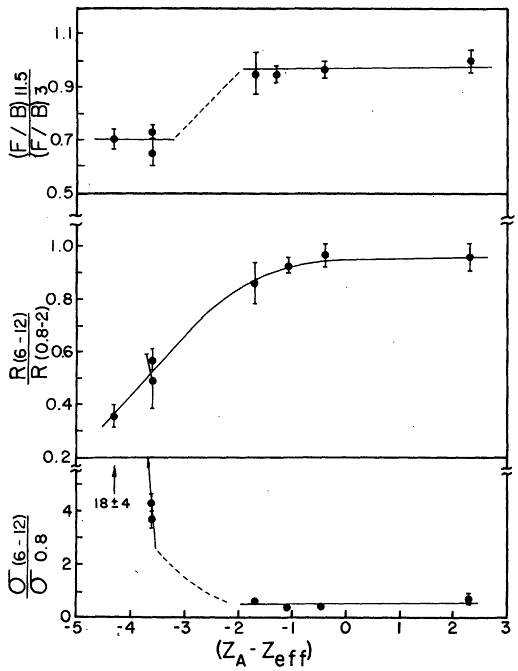

FIG. 3. Dependence on composition of the ratio of cross sections at 6–12 to 0.8 GeV (bottom panel), of the ratio of ranges at 6–12 to 0.8–2 GeV (middle), and of the ratio of F/B values at 11.5 to 3 GeV (top). The numerical ratio in the bottom panel refers to ^{131}Ce.

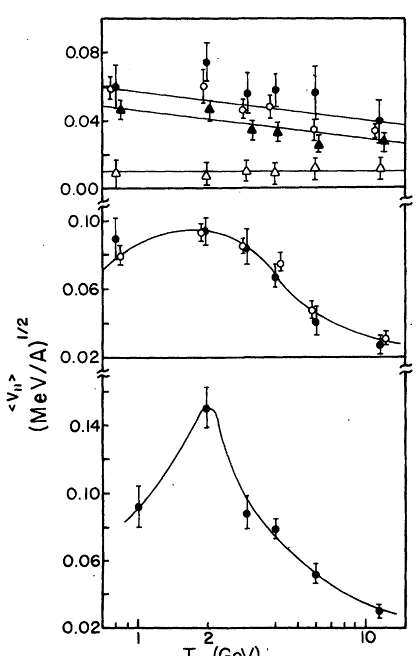

FIG. 4. Energy dependence of $\langle v_{\parallel} \rangle$ of struck nuclei leading to formation of observed products. Bottom panel, ^{131}Ce; middle, ^{131}La(●), ^{128}Ba(○); top, ^{131}Ba(●), ^{133}Ba^m(○), ^{135}Ba^m(▲), ^{140}Ba(△). The curves show the trends in the data.

of $\langle v_{\parallel} \rangle$ with composition, depicted in Fig. 5, indicates that at low energies this quantity exhibits a fairly strong inverse dependence on $(Z_A - Z_{\text{eff}})$. This correlation gradually weakens with increasing proton energy and at 11.5 GeV the $\langle v_{\parallel} \rangle$ are virtually independent of product composition.

The trends exhibited by the $\langle v_{\parallel} \rangle$ values at the lower energies are readily understandable in terms of the two-step model. Since in this energy regime $\langle v_{\parallel} \rangle$ is proportional to the mean excitation energy of the struck nuclei, $\overline{E}^*$,[36,37] the above trends are also followed by this quantity. The increase in $\overline{E}^*$ with proton energy observed for the most neutron-deficient products is thus an indication that these nuclei are only formed with appreciable probability in high-energy transfer processes. As indicated above, this behavior is consistent with the rising excitation functions of these products. Similarly, the nearly constant $\overline{E}^*$ obtained for the less neutron-deficient nuclides are consistent with their falling excitation functions. The increase in $\overline{E}^*$ with increasing neutron deficiency that can be inferred from the behavior of the $\langle v_{\parallel} \rangle$ values is also consistent with the two-step

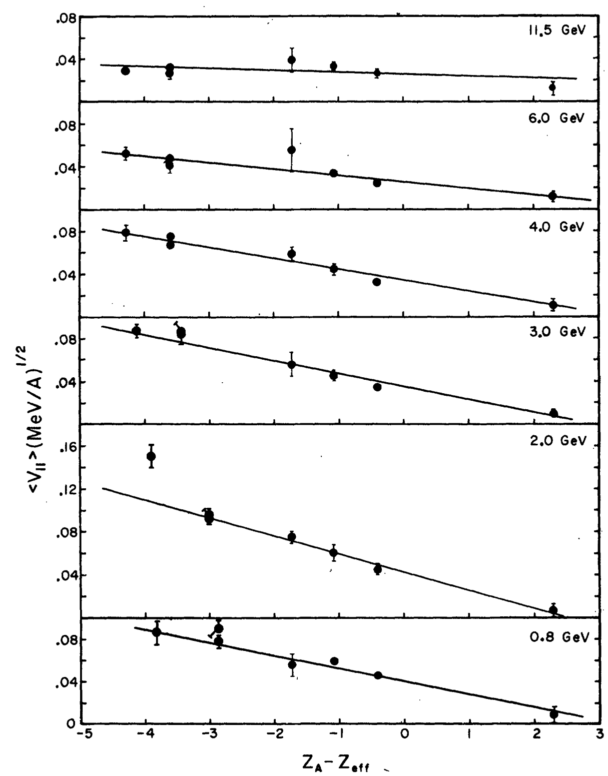

FIG. 5. Dependence of $\langle v_{\parallel} \rangle$ on product composition at various proton energies. The $(Z_A - Z_{\text{eff}})$ of cumulative products below 11.5 GeV were estimated on the basis of the cross sections of isobaric nuclides. The lines show the trends in the data.

model and has, in fact, been thoroughly investigated and explained.[38]

The behavior of the $\langle v_{\parallel} \rangle$ values associated with the formation of the most neutron-deficient products at the higher energies is not readily explainable. As has been pointed out before,[1,2] the combination of decreasing $\bar{E}^*$ and a rising excitation function is highly implausible. The observed sharp decrease in the $\langle v_{\parallel} \rangle$ values is thus an indication of a breakdown in the relation between momentum and energy transfer at high excitation energies, or of the inapplicability of the two-step model to these processes. In any case, the $\langle v_{\parallel} \rangle$ values are consistent with the other data in indicating that a profound change in the nature of highly inelastic interactions of protons with heavy elements occurs in the vicinity of 3 GeV.

B. Explanation of results in terms of a change in the nature of high-energy proton-nucleus interactions

A variety of explanations for at least some of the changes that occur at ~3 GeV have been proposed. These have been conveniently summarized by Kaufman, Steinberg, and Weisfield.[18] We wish to expound here the role played by the change in the nature of hadron-nucleus interactions at high energies.

Various experiments performed in recent years have indicated that the conventional intranuclear cascade model of high-energy reactions, which is in generally satisfactory agreement with experiment up to incident hadron energies of 1–2 GeV,[39] cannot be valid at much higher energies. It is thus found that the ratio of the mean number of relativistic charged particles emitted in hadron-nucleus collisions to that emitted in hadron-proton collisions, usually designated R_A, is nearly independent of energy and target A in the high-energy regime.[40] Moreover, the values of R_A tend to be rather small, not exceeding a value of ~2.2 for the heaviest elements. If the relativistic secondary particles created in hadron-nucleon collisions at high energies were to participate in an intranuclear cascade, R_A would increase sharply with target A. Since this effect is not seen it has been concluded that the fast secondary particles do not cascade. This result has been explained by a number of models such as the energy flux model,[41] the collective tube model,[42] and the effective target model.[43] Although these models are conceptually different, they all stress the role of relativistic effects at high energies. A fully relativistic proton thus sees a target nucleus that is Lorentz contracted to a narrow disk. The nucleons that lie in the path of the proton consequently constitute a contracted array and interact coherently with it. This is in marked contrast to the situation at lower energies, where the interaction of the projectile consists of a series of collisions with individual quasifree nucleons. The collective interaction gives rise to a highly excited state of hadronic matter. Owing to relativistic time dilation, this state does not decay to the observed ensemble of energetic secondary particles until it is well outside the struck nucleus. As a result, the secondaries do not participate in the intranuclear cascade.

While the above models have been proposed to explain the results of hadron-nucleus interactions at Fermilab energies, they may already be applicable at energies as low as 5–10 GeV. An estimate of the lower limit of their validity can be obtained on the basis of some simple considerations. In order for the hadronic flux to be ejected from the nucleus prior to decay, its intrinsic lifetime τ_0 must be sufficiently dilated to permit escape, i.e., $d \leq c\tau_0\gamma$, where d is the diameter of the target nucleus and $\gamma = (1-\beta^2)^{-1/2}$. We have estimated the minimum proton energy consistent with this condition as ~8 GeV. This result is based on $\tau_0 = 5\times10^{-24}$ sec, consistent with $\Gamma \sim 120$ MeV for various baryon states,[44] and $d = 1.3\times10^{-12}$ cm for ^{238}U (Ref. 45).

More direct evidence on the importance of coherent interactions at low energies has been presented in a recent analysis of the production of heavy particles at energies below their threshold for production in nucleon-nucleon collisions.[46] While such particles can be produced in subthreshold interactions between individual nucleons on account of Fermi motion, they can be made in much higher yield in collective interactions because of the greater energy available for particle creation. A comparison of these two models with the excitation function for $\bar{p}$ production in the interaction of copper with 3–6 GeV protons indicates that a substantial contribution of a collective mechanism is needed to account for the relatively high $\bar{p}$ yields.[46] Additional experimental evidence bearing on the importance of coherent interactions at low energies has been presented by Gutay *et al.*[47]

A coherent interaction model coupled with some additional assumptions appears to be capable of explaining many of the results of present interest. The rapid ejection of a tube of nuclear matter leaves behind a residual nucleus with a "hole" punched out along the beam direction. This residue is nearly stationary in the laboratory system since the momentum of the incident proton is carried off by the ejected ensemble. We assume that this unstable residue breaks up into two or more fragments, as shown schematically in Fig. 6. Because the hadronic flux is ejected along the beam direction, Coulombic repulsion causes the fragments to separate along a trajectory that is transverse to that of the beam.[48] The observed products, which result from the deexcitation of these fragments, will thus have low F/B and low associated $\langle v_{\parallel} \rangle$ values as well as sideward-peaked angular distributions. These are precisely the results obtained at 11.5 GeV and above. While it does not seem unreasonable to suppose that a bead-shaped nucleus should undergo a rapid two-body breakup, we offer no justification for this assumption other than its ability to account for the data. The calculation of the dynamics of this process will obviously be a major task.

It should be pointed out that the peak in F/B observed at 3 GeV does not actually require that the transition from a cascade to a coherent interaction mechanism occur at this low an energy. According to the cascade model, F/B should increase with bombarding energy as long as the deposition energy required to form the product in question increases. Once the optimum $\bar{E}^*$ is attained, the forward momentum imparted to the struck nucleus becomes proportional to the ratio of the momentum of the incident proton to its kinetic energy. At nonrelativistic energies this ratio varies as $T_p^{-1/2}$ and so F/B, which is a measure of the forward momentum of the struck nucleus, initially decreases with increasing T_p. However, at fully relativistic energies the above ratio becomes independent of T_p and F/B must level off. The published data on F/B of ^{131}Ba and other products have been analyzed in this fashion by Scheidemann and Porile.[17] It was found that the cascade model

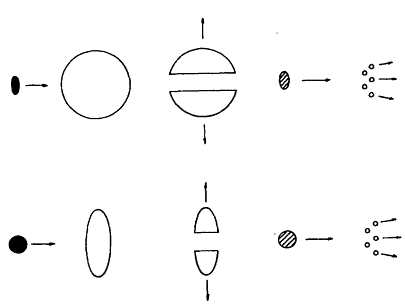

FIG. 6. Schematic representation of collective interaction model of hadron-nucleus collisions at high energies. A central collision leading to a fast two-body breakup is depicted in the laboratory system (top) and the projectile frame (bottom).

could match the initial decrease in F/B up to approximately 4–6 GeV. It is the continuing decrease in F/B observed at higher energies that is inconsistent with the cascade model. As indicated above, this decrease occurs in an energy regime where a collective interaction model may be physically plausible.

Two additional points require consideration. First, we address the question of the identity of the products formed in the breakup of the residue. We believe that the impact parameter of the initial collision is of importance in this connection. A central collision, such as that depicted in Fig. 6, might indeed lead to a fast two-body breakup process as indicated. Considerable particle evaporation would presumably accompany this process because of the excess surface energy of the residue,[49] the possible action of frictional forces between the hadronic flux and the residue,[50] and the occurrence of final state interactions.[51] The end result would be the formation of moderately light fragments. As an example, we cite the case of Sc nuclides, whose excitation functions increase sharply with proton energy up to ~10 GeV, indicating that they are formed in a highly inelastic process.[17] It has been found[52] that the spectra of these fragments are reasonably narrow at high energies, suggesting that a two-body breakup process is plausible. A collision at intermediate impact parameters would, in this view, lead to deep spallation products such as ^{128}Ba. While a collective interaction would still occur, the residual nucleus would not necessarily be cleaved into just two fragments but would instead break up into several very light fragments and nucleons, leaving a massive residue to decay to the observed product. Finally, peripheral collisions would not involve collective interactions since the amount of nuclear matter in the path of the incident proton is too small. Instead, the cascade model should retain its applicability. According to this model, interactions of this type involve relatively low energy transfers and so result in the formation of spallation and binary fission products. Since there is no change in the nature of the primary interaction, the F/B ratios of these products do not peak at 3 GeV and their angular distributions remain essentially unchanged. The formation of ^{140}Ba appears to be a good example of this process.

Second, we address the question of the observed decrease of the ranges in the context of the proposed model. As indicated above, much of the range decrease occurs below 3 GeV, where the data are still consistent with the cascade mechanism. It is known[8,9,21] that the drop in the mean range of Ba nuclides is accompanied by a change in the range or energy spectrum from narrow and symmetric to broad and asymmetric. This is an indication that the mechanism changes from binary fission to deep spallation. It is certainly tempting to ascribe this change to the transition from nucleonic to collective interactions since it is difficult to conceive that two such profound changes can occur within an interval of a few GeV and be unrelated. However, the possibility cannot be excluded solely on this basis.

It must be mentioned, in this connection, that a two-body breakup process of the type depicted in Fig. 6 involves the formation of two fragments in rather close proximity to each other. This would tend to lead to high kinetic energies rather than to the observed low energies, unless other factors are also operative. We believe that the extensive mass (and charge) loss resulting from the factors referred to above may be responsible.

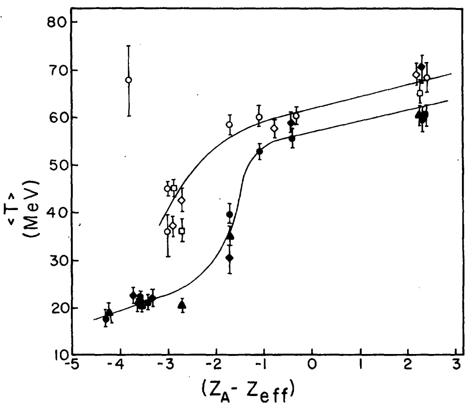

FIG. 7. Mean kinetic energies of Ba and $A=131$ nuclides at 11.5 GeV (closed points) and 2 GeV (open points). The curves show the trends in the data. ○, present results; △, Refs. 11 and 15; □, Ref. 1; ◇, Ref. 21 (11.5 GeV) and Ref. 9 (2 GeV).

C. The problem of ^{131}Ba

In previous publications from our laboratory[11,21] it was established that the range of ^{131}Ba (I) formed in the interaction of ^{238}U with 11.5-GeV protons is longer than those of the neighboring deep spallation products but shorter than those of nearby fission products. A decomposition of the momentum spectrum derived from the differential range in fact indicated that deep spallation accounted for $75 \pm 16\%$ of the observed yield.[21] As suggested in the preceding sections, the occurrence of a peak in F/B and the abrupt decrease in range near 3 GeV constitute a signature of the transition between binary fission and deep spallation and suggest that there is a concomitant change from a cascade to a collective interaction mechanism. However, Fig. 2 indicates that the data for ^{131}Ba display neither one of these features. The question thus arises as to whether the present results for this nuclide are in disagreement with the previous reports.[11,21] A detailed comparison is clearly in order.

The mean kinetic energies derived from the various integral and differential range experiments are displayed as a function of $(Z_A - Z_{eff})$ in Fig. 7. The ranges listed in Table III were converted to energies by means of a procedure described in detail elsewhere.[17] It is seen that the present results are in generally good agreement with the results of previous measurements at 11.5 GeV.[1,11,15,21] In particular, the kinetic energy of ^{131}Ba, though slightly higher, is in accord with previous determinations and does indeed fall between the values of the deep spallation and fission products.

Figure 7 also displays the kinetic energies derived from the 2-GeV data. Here too, our results agree with previous determinations[1,8,9] with the exception of ^{131}Ce, whose kinetic energy is much higher than expected from the trend established by the more neutron-rich isotopes, and appears to be anomalous. At this lower proton energy, the kinetic energies do not divide into two distinct groups since all the products appear to be primarily the result of binary fission.[8] Only products with $(Z_A - Z_{eff}) < -2.5$ have significantly lower energies. This difference reflects the transition from fission to deep spallation that occurs in this energy regime. It has thus been estimated[8] that the deep spallation contribution to the formation of ^{131}Ba (C) $(Z_A - Z_{eff} = -2.7)$ is ~18%. On the other hand, the present results for ^{131}Ba (I) $(Z_A - Z_{eff} = -1.7)$ follow the trend established by the fission products at this energy.

The present results thus indicate that the properties of products whose composition places them between the fission and deep spallation regions are complex and not readily understood. If only the 11.5-GeV data[11,21] are examined, the magnitude of the mean energy of ^{131}Ba and the width of its distribution suggest that this nuclide is largely formed in deep spallation. On the other hand, the energy dependence of the recoil properties as well as the shape of the excitation function are very similar to those of the more neutron-rich fission products and not at all like those of the deep spallation products. It is, of course, possible that the experimental uncertainties mask the smaller effects that would be expected from a combination of the two mechanisms but this does not seem very likely. Contributions from still other processes may also be possible. It is interesting to note in this connection that Starzyk and Sugarman[13] postulated the occurrence at high energies of a fission process involving moderately high deposition energies. This so-called Fission II process was invoked to account for the mismatch between the $(Z_A - Z_{eff})$ values corresponding to the transition between products having long and short ranges and that at which a minimum in the charge dispersion is observed. Interestingly enough, this process is expected to make its biggest contribution at $A=131$ in the vicinity of ^{131}Ba.[13] Because of the higher excitation energies imparted to the struck nucleus, the products would have lower kinetic energies and ranges than the more neutron-rich fission products. Our results do not, of course, prove that a distinctive process of this type in fact occurs. We merely cite this mechanism as an indication of the additional complexities that are possible. It would certainly be of interest to perform careful measurements on other products with similar composition to ascertain whether the results for ^{131}Ba constitute more than just an isolated anomaly.

V. CONCLUSIONS

A study of the energy dependence of the recoil properties and cross sections for the formation of several isobaric nuclides with $A=131$ as well as some neighboring Ba nuclides in the interaction of ^{238}U with high-energy protons reveals a striking dichotomy based on product composition: Highly neutron-deficient nuclides $(Z_A - Z_{eff} \leq -3.6)$ have rising excitation functions up to 5 or 6 GeV, F/B ratios that peak at 3 GeV, and ranges that decrease abruptly somewhat below this energy. On the other hand, neutron-rich and moderately neutron-deficient nuclides $(Z_A - Z_{eff} \geq -1.7)$ have falling excitation functions, and F/B ratios and ranges that decrease slowly and featurelessly with increasing proton energy.

Two major changes in the nature of the reaction mechanism have been invoked to explain the results obtained for the neutron-deficient products. The decrease in the ranges and the concomitant broadening of the spectra[21] can be satisfactorily explained as arising from a transition between binary fission and deep spallation. The peak in F/B and the associated change in the angular distributions from forward to sideward peaked[19-21] are explainable as the result of a fundamental change in the nature of near-central proton-nucleus collisions at fully relativistic energies coupled with some additional assumptions about the breakup modes of the residual nucleus. Owing to the Lorentz contraction of the nucleus as viewed by the projectile, the latter interacts collectively with the ensemble of nucleons lying in its path. Because of time dilation, the ejected matter does not decay to its final multiparticle state until it is outside the nucleus. As a result of this initial interaction the residual nucleus has a hole punched out along the beam axis and fragments in a transverse direction. While collective interaction models have been advanced to explain the results of many experiments on hadron-nucleus interactions above 100 GeV, they appear to be valid even in the regime of present interest. The transition between the conventional intranuclear cascade consisting of a series of individual nucleon-nucleon collisions and a collective proton-effective target interaction may thus occur, at least for central collisions, at energies below 10 GeV. Although the products whose recoil properties are consistent with the above mechanism at 11.5 GeV constitute only ~30% of the isobaric yield at $A=131$,[11] the mechanism is also expected to be of importance in the formation of lighter fragments. Collective interactions followed by breakup of the residue may thus account for a sizable fraction of the total reaction cross section of heavy elements at high energies. The incorporation of these features into a quantitative model of hadron-nucleus interactions is posed as a challenge to the theorists.

The necessity of separately invoking a change from fission to deep spallation as well as one from nucleonic to collective interactions within an energy interval of at most a few GeV appears to be unduly complex. It would be an esthetically pleasing simplification if the first of these changes were to be just another manifestation of the second. However, more detailed exclusive experiments are needed to establish this point.

Our model is subject to a number of relatively simple experimental verifications. Since products that appear to be associated with the more peripheral interactions do not exhibit the characteristic peak in F/B and change in the angular distribution, we should not expect to see these effects for targets of sufficiently light elements to obey the condition that the number of nucleons in the path of an incident proton along a central trajectory be no larger than that encountered in a peripheral interaction with a heavy element. In a different vein, since less massive hadrons than the nucleon become fully relativistic at lower energies, the effects in question should be observable at lower bombarding energies for lighter projectiles. A study of the interaction of heavy elements with pions would be informative in this respect.

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