Recoil properties of ^{24}Na and ^{28}Mg formed by the interaction of ^{197}Au and ^{238}U with 3–300-GeV protons*

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The recoil properties of ^{24}Na and ^{28}Mg formed by the interaction of protons of energy 3.0–300 GeV with targets of gold and uranium have been measured by a thick-target technique. γ-ray spectroscopy without chemical separation was used to measure the nuclides of interest. The forward-to-backward ratio (F/B) of both nuclides from both targets exhibits a maximum near 3 GeV, decreasing from a value of about 2.0 at that energy to about 1.3 at 300 GeV. This behavior is strikingly similar to that of several neutron-deficient medium-mass nuclides formed from uranium, such as ^{131}Ba, and seems to indicate that both types of products are formed in similar processes. The mean ranges of the light nuclides were found to decrease by about 25% from 3 to 300 GeV, also similar to the behavior of the neutron deficient products, although smaller in magnitude.

[NUCLEAR REACTIONS ^{197}Au and ^{238}U (p, fragmentation) ^{24}Na and ^{28}Mg, E = 3–300 GeV; measured thick-target recoil properties.]

I. INTRODUCTION

The formation of relatively light nuclei ($A \lesssim 30$) in the interaction of energetic protons with heavy targets has been considered to occur by a characteristic mechanism, *fragmentation*. This is a nuclear process occurring on a fast time scale, comparable to that of the intranuclear cascade, and involving excitation energies of the order of 0.5 GeV or more. The evidence for such a process comes from a number of sources, including excitation functions, energy and angular distributions, and charge distributions.[1] The energetic threshold for production of these light nuclei from heavy targets is several hundred MeV and their excitation functions rise steeply, only leveling off above several GeV bombarding energy. Examples of such excitation functions include the formation of ^{18}F, ^{24}Na, ^{28}Mg, and ^{32}P from lead,[2] ^{7}Be from silver and gold,[3] ^{32}P from holmium, gold, and thorium,[4] and ^{24}Na and ^{28}Mg from copper, silver gold, and uranium.[5] In the latter work[5] the excitation functions for protons and helium ions were compared, with the conclusion that formation of ^{24}Na required a mean deposition energy of 0.5–0.7 GeV, nearly independent of target and projectile.

Further evidence on the nature of fragmentation is provided by measurements of kinetic energy spectra and angular distributions of the fragments. Using a thin-target, thin-catcher recoil technique Cumming *et al.*[6] measured the angular distribution and the energy spectra at three angles of ^{24}Na formed by bombardment of Bi with 2.9-GeV protons. Analysis of the data showed that there was no moving system in which both the angular distribution and velocity spectra are symmetric about the direction perpendicular to the beam. Thus the cascade-evaporation model, in which the reaction is assumed to occur in two distinct steps, (the second step having no memory of the first), cannot account for the data. Instead, it was concluded that the breakup of the excited cascade residue into fragments ultimately leading to ^{24}Na occurred in a time comparable to that of the cascade itself, so that the fragments retained some memory of the beam direction.

More detailed energy and angular distributions of a large number of light nuclei formed from 5.0–5.5-GeV proton bombardment of uranium and silver have been recently measured.[7–9] Individual nuclides were identified and their energies measured at different angles with semiconductor detectors using the ΔE-E technique. These results essentially confirmed the conclusions based on the ^{24}Na recoil measurements, that a substantial fraction of the fragments are formed in a fast nonequilibrium mechanism.

Fragmentation has also been postulated[10–12] as a mechanism for the formation of neutron-deficient medium-mass nuclides ($60 \lesssim A \lesssim 150$) from heavy targets, particularly uranium in the multi-GeV region. Such nuclides may be considered to be the complementary products of the light fragments, formed as the result of a long evaporation chain after the initial fragmentation. The isobaric yield patterns of medium-mass nuclides formed from uranium exhibit a split into separate neutron-deficient and neutron-excess peaks as the bombarding

energy increases from 0.5 to 1.0 GeV,[13] indicating the energy region in which a new mechanism begins to contribute. Recoil measurements on a number of these nuclides have shown that the mean kinetic energy of a typical neutron-deficient nuclide is approximately half as large at bombarding energies above a few GeV as it is at energies below 1 GeV, while the mean kinetic energy of a typical neutron-rich nuclide is nearly independent of bombarding energy. Among the nuclides showing this behavior from uranium targets are isotopes of iodine[10,14] and antimony,[15] and isobaric products near mass numbers $A=111$ [16] and $A=131$.[17] This decrease in mean kinetic energy has been shown to occur between proton energies of 1.0 and 5.0 GeV for the neutron-deficient nuclides ^{83}Sr and ^{131}Ba.[12]

The decrease in mean kinetic energy of neutron-deficient nuclides has been interpreted as follows. Below 1 GeV the main process occurring is binary fission of the excited cascade residue, occurring as a slow second step. Neutron evaporation may occur before and after the fission, and the more highly excited the cascade residue is the more neutron deficient the final products will be. Recoil measurements of a number of products from 0.45-GeV proton bombardment of uranium[18,19] confirm this view, showing that mean kinetic energies are consistent with such a binary fission process. At proton energies above 1 GeV, where the formation cross sections of the light fragments are rising rapidly, it has been suggested[10,12] that neutron-deficient nuclides are formed to an increasing extent by fragmentation. This accounts for the decrease in mean kinetic energy in two ways: The Coulomb repulsion of the two fragments is lower than in the case of the predominantly symmetric fission process, and the forward emission of the light fragment causes a backward recoil of the heavy one, thus lowering its (net forward) momentum.

The energy dependence of the recoil properties of several medium-mass nuclides formed from uranium has been studied over the energy range 0.45–11.5 GeV,[12] and extended recently to 300-GeV energy.[20,21] In the same region that the mean ranges of ^{83}Sr and ^{131}Ba decrease, 1–5 GeV, the forward-to-backward ratio (F/B) showed a remarkable behavior, first increasing from the value found at lower energies, going through a maximum near 3 GeV, and then decreasing again. In contrast, the mean ranges and F/B values of the neutron-rich nuclides ^{91}Sr and ^{140}Ba were nearly constant over the entire energy range. This peak in F/B would seem to be another characteristic phenomenon, in addition to the range decrease, by which one may identify those nuclides whose formation mechanism is changing.

Since the recoil properties of light fragments have not been studied over a large range of incident energy we have undertaken such a study. Data on the nuclides ^{24}Na and ^{28}Mg formed from several targets at proton energies of 0.7 and 3.0 GeV are in the literature,[5] and we have extended these measurements up to 300 GeV in the present work.

II. EXPERIMENTAL PROCEDURE

Irradiations were done in both the internal circulating beam and the external beam at the Argonne National Laboratory zero gradient (ZGS) proton synchrotron ($E_p=3.0$–11.5 GeV), in the external beam at the Brookhaven National Laboratory alternating gradient synchrotron (AGS) ($E_p=28$ GeV), and in the external beams at the Fermi National Accelerator Laboratory ($E_p=200$–300 GeV). The internal beam irradiations were done at 3.0 GeV (gold target) and at 6.0 GeV (uranium target), all of the remainder being done externally. Targets consisted of metallic foils, approximately 25 mg/cm^2 thick, sandwiched between Mylar catcher and guard foils, approximately 18 mg/cm^2 thick, all foils being thicker than the mean range of the products of interest. The uranium foils were of depleted ^{238}U and were cleaned of their oxide coating with dilute HNO_3 before each bombardment.

In the external beam experiments there were three Mylar foils on each side of the target, the second and third foils acting as activation blanks and guards. This target stack was sealed in a polyethylene bag under vacuum, so that atmospheric pressure kept the foils in close contact and they were protected from contamination. Irradiations lasted from 2 to 48 h, depending on beam intensity and on access to the target area. Since only activity ratios of an individual radionuclide in the target and catchers are needed, there was no need to correct for saturation or variation of beam intensity during the irradiation. After the irradiation an area including all of the beam spot was cut out of the stack for counting. For the internal ZGS bombardments only three Mylar foils were used, in order to maximize the number of traversals of the target by the circulating beam, thus providing only one activation blank. The Mylar foils extended out about 1 mm further than the target in order to collect all recoils escaping from the leading edge.

The activation-blank Mylar foils normally had a small amount of ^{24}Na activity but showed no detectable ^{28}Mg. The ^{24}Na activity was the same in all of the blanks, indicating that it was due to an impurity in the Mylar. The absence of any ^{28}Mg indicates that there is no appreciable fraction of recoils with high enough kinetic energy

TABLE I. Decay properties of observed radionuclides. Data taken from the compilation of W. W. Bowman and K. W. MacMurdo, At. Data Nucl. Data Tables 13, 90 (1974).

Nuclide	Half-life	γ-ray energy (keV)	Abundance (%)
^{24}Na	15.0 h	1368.5	100.0
		2753.9	99.9
^{28}Mg	21.1 h	941.7	36
		1342.2	54
		1778.9 [a]	100.0

[a] Daughter ^{28}Al activity.

($\gtrsim$180 MeV) to penetrate 18 mg/cm^2 of Mylar.

The target and catcher foils were counted directly, without chemical separations, with a spectrometer consisting of a Ge(Li) detector of about 2.0-keV resolution at 1330 keV and a 4096-channel analyzer with magnetic tape readout. The resulting γ-ray spectra recorded as a function of time for each foil were analyzed with the aid of the computer program SAMPO.[22] The decay properties of ^{24}Na and ^{28}Mg are summarized in Table I, and portions of the observed spectra in the regions of the listed γ rays are shown in Fig. 1, for uranium, and Fig. 2, for gold. In Figs. 1 and 2 the upper spectrum is that of the target and the lower is that of the forward catcher, both recorded approximately two days after the end of the bombardment.

Figures 1 and 2 illustrate the general aspects of the γ-ray spectra of heavy targets and their recoil catchers after bombardment with energetic protons. The catcher spectra are less complex than those of the targets because many of the products have low recoil ranges, especially those closest in mass to the target. However, the fragmentation products, such as ^{24}Na and ^{28}Mg, have comparatively large recoil ranges, so that they are prominent in the catcher spectra, and in addition the energetic γ rays of these two nuclides make them especially prominent when compared to nuclides of similar half-life.

A close examination of Figs. 1 and 2 reveals the similarities and differences between the two targets. The ^{28}Mg 1342.2-keV γ ray exhibits a low-energy shoulder in both target spectra which is absent in the catcher spectra. The shoulder is probably due to an interfering nuclide of low recoil range whose half-life appears to be comparable to that of ^{28}Mg. The interference makes this γ ray unsuitable for ^{28}Mg analysis in the targets. There are several interfering γ rays in the vicinity of the ^{24}Na 1368.5-keV line in the targets, but the computer analysis was able to resolve this structure satisfactorily. There is a low-intensity shoulder on the ^{24}Na 2753.9-keV line in both target spectra, but it is more prominent in gold. This latter interference is tentatively identified as the 2748.2-keV γ ray in 52-h ^{170}Lu. The ^{28}Mg 941.7-keV line is resolvable in the uranium spectra but is almost

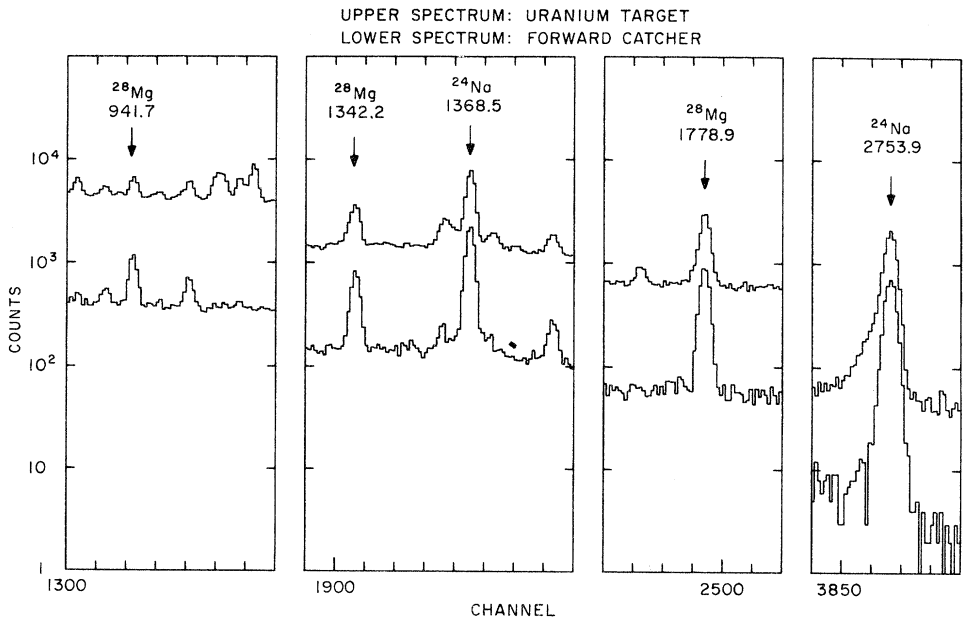

FIG. 1. Portions of the γ-ray spectra of the uranium target (upper spectrum) and the forward recoil catcher (lower spectrum) in the vicinity of the characteristic γ rays of ^{24}Na and ^{28}Mg. The spectra were taken approximately 2 days after bombardment with 11.5-GeV protons.

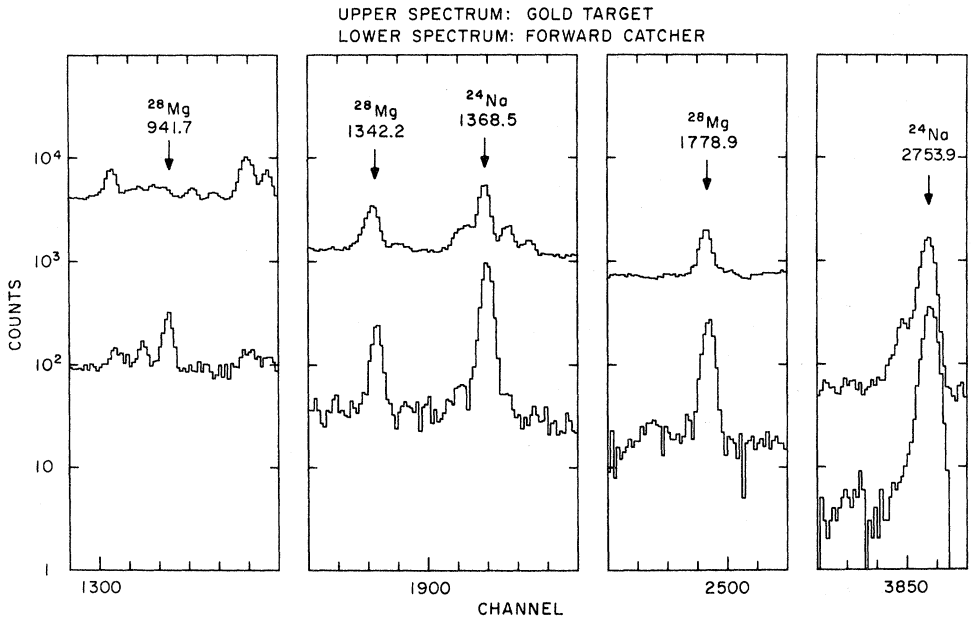

FIG. 2. Portions of the γ-ray spectra of the gold target (upper spectrum) and the forward recoil catcher (lower spectrum) in the vicinity of the characteristic γ rays of ^{24}Na and ^{28}Mg. The spectra were taken approximately 2 days after bombardment with 11.5-GeV protons.

invisible in the gold spectra.

In the spectral analysis of the catcher foils all of the γ rays listed in Table I were used, and the decay curves showed a single component of the correct half-life. Furthermore, the relative intensities of the γ rays were in agreement with the literature values. For the uranium targets four of the listed γ rays could be analyzed satisfactorily, the exception being the ^{28}Mg 1342.2-keV line, while for the gold targets both that and the 941.7-keV γ rays could not be used.

The quantities determined from this analysis are F and B, the fractions of activity of a given nuclide recoiling out of the target in the forward and backward directions, respectively. These quantities depend on the analysis of both catcher

TABLE II. Recoil properties of observed nuclides.

E_p (GeV)	No. of determinations	^{24}Na F/B	^{24}Na $2W(F+B)$ (mg/cm^2)	^{28}Mg F/B	^{28}Mg $2W(F+B)$ (mg/cm^2)
		Uranium target			
0.70 [a]		1.64± 0.04	13.4± 0.4	1.52± 0.07	17.3± 0.6
3.0 [a]		2.06± 0.05	16.2± 0.4	1.86± 0.09	18.0± 0.8
6.0	1	1.97± 0.05	15.3± 0.5	1.80± 0.05	16.7± 0.6
11.5	2	1.62± 0.04	14.3± 0.5	1.51± 0.04	15.7± 0.6
300	2	1.33± 0.03	12.8± 0.4	1.29± 0.04	14.4± 0.5
		Gold target			
0.70 [a]		1.68± 0.04	15.1± 0.4	1.53± 0.07	18.1± 0.8
3.0 [a]		2.09± 0.05	13.5± 0.4	2.05± 0.10	15.4± 0.6
3.0	1	2.02± 0.06	15.5± 0.8	1.91± 0.06	17.8± 1.0
6.0	1	1.75± 0.05	13.3± 0.5	1.62± 0.10	14.5± 0.5
11.5	3	1.53± 0.03	12.7± 0.4	1.44± 0.04	14.2± 0.4
28	2	1.37± 0.03	11.8± 0.4	1.31± 0.04	13.2± 0.5
200	1	1.31± 0.05	11.2± 0.5	1.26± 0.05	12.7± 0.5
300	2	1.31± 0.03	11.8± 0.4	1.25± 0.04	13.2± 0.4

[a] Data from Ref. 5.

and target spectra, but the ratio F/B depends only on the catcher analysis. From the discussion above it follows that F and B for ^{28}Mg from gold depend on the analysis of only the 1778.9-keV line, while for the same nuclide from uranium and for ^{24}Na for both targets two γ rays can be used. Hence these quantities are subject to more uncertainty for ^{28}Mg from gold. However, the F/B ratio for ^{28}Mg can be determined using three γ rays and is thus less subject to error. In addition, since the catcher spectra are simpler than the target spectra their analyses are more precise, which also makes the F/B ratio more accurate than either quantity alone. These considerations enter into the errors assigned to the experimental results.

III. RESULTS AND DISCUSSION

The recoil parameters obtained in these experiments are the forward-to-backward ratio, F/B, and the range, $2W(F+B)$, where W is the target thickness in mg/cm^2. [The mean range of the recoils is somewhat smaller[18] than $2W(F+B)$, but it is conventional to refer to the latter quantity as range.] These recoil parameters for ^{24}Na and ^{28}Mg formed from targets of uranium and gold are summarized in Table II, which also gives the values measured by Crespo *et al.*[5]

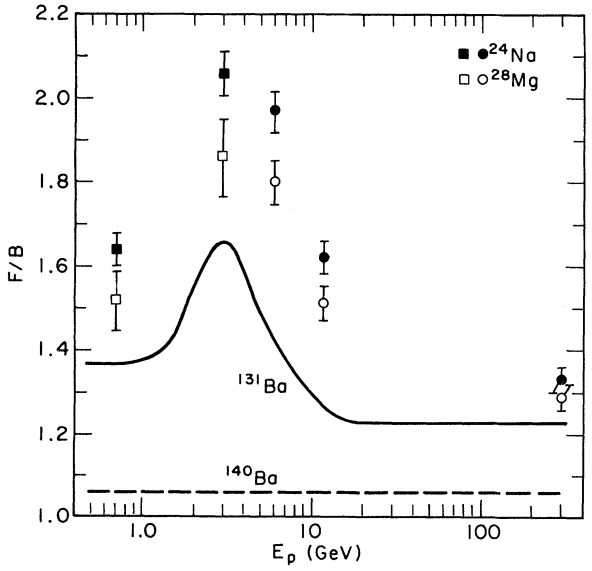

FIG. 3. Energy dependence of the forward-to-backward ratio (F/B) for ^{24}Na and ^{28}Mg formed from uranium. The points at 0.7 and 3.0 GeV are from Ref. 5. Smooth curves show the energy dependence of F/B for ^{131}Ba and ^{140}Ba formed from uranium (Refs. 12, 20, and 21) for comparison.

Corrections for scattering and edge effects[23] have not been made to the experimental data. The former correction results from preferential scattering of heavy recoils near the end of their range into a low-Z material (e.g., Mylar) from a high-Z material (U, Au). The correction amounts to 3–4% for fission fragments[23] but is probably smaller for the lighter recoils measured here.[5] Such a correction, if made, would lower all ranges by the same percentage and leave the F/B ratios unchanged. The edge correction[23] is absent for all of the external beam bombardments, since there is no edge exposed to the beam, and would be about 0.5% for the target thicknesses used in the internal bombardments.

The errors given in Table II represent estimates of the standard deviation, based on the agreement among the replicate determinations using different γ rays and on replicate bombardments. The agreement among different γ rays in the same bombardment was usually within the limits given by the least-square analyses of the various decay curves, and the agreement of the F/B values between replicate bombardments at the same energy was also satisfactory. The agreement of the values of $2W(F+B)$ between replicate bombardments was somewhat poorer, probably due to the difficulties in analyzing the complex spectra of the targets, and the errors assigned to these values reflect such uncertainties.

The data in Table II are presented graphically in Figs. 3–6. Figure 3 shows the values of F/B

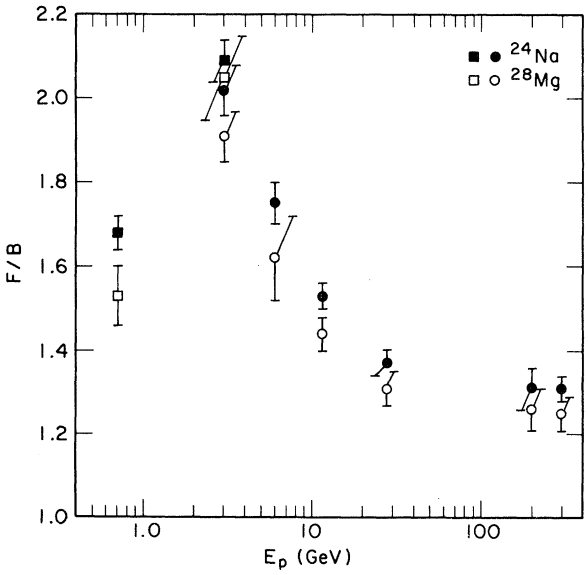

FIG. 4. Energy dependence of the forward-to-backward ratio (F/B) for ^{24}Na and ^{28}Mg formed from gold. (■, □): Ref. 5. (●, ○): this work.

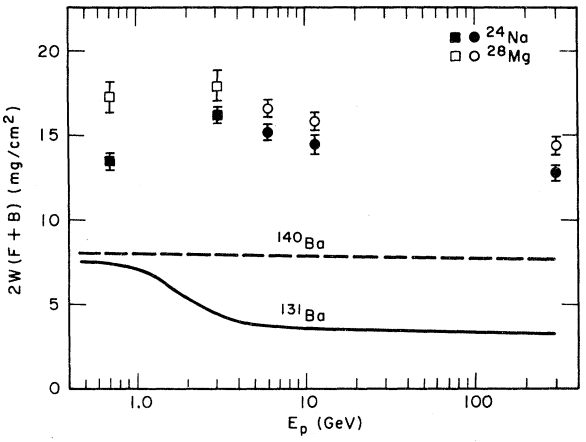

FIG. 5. Energy dependence of the mean range $[2W(F+B)]$ for ^{24}Na and ^{28}Mg formed from uranium. (■, □): Ref. 5. (●, ○): this work. Smooth curves show the energy dependence of $2W(F+B)$ for ^{131}Ba and ^{140}Ba formed from uranium (Refs. 12, 20, and 21) for comparison.

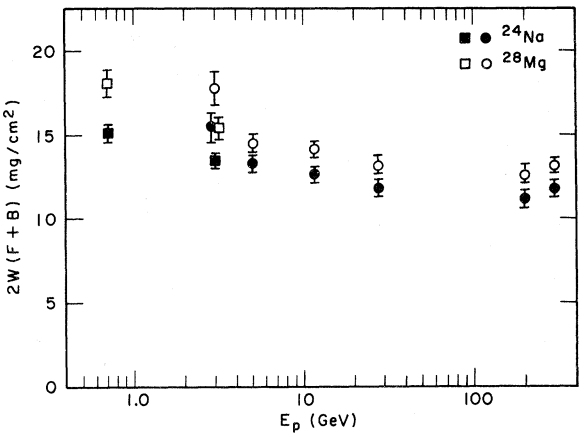

FIG. 6. Energy dependence of the mean range $[2W(F+B)]$ for ^{24}Na and ^{28}Mg formed from gold. (■, □): Ref. 5. (●, ○): this work.

for ^{24}Na and ^{28}Mg from uranium and Fig. 4 these values from gold. Figures 5 and 6 show the values of $2W(F+B)$ for the uranium and gold targets, respectively. In addition Figs. 3 and 5 show as smooth curves the behavior of F/B and $2W(F+B)$ for typical neutron-deficient (^{131}Ba) and neutron-rich (^{140}Ba) nuclides formed from uranium[12,20,21] for comparison with the present results. The recoil properties of ^{24}Na and ^{28}Mg from uranium and gold are very similar; in both cases the values of F/B peak at about 3 GeV with a value of about 2.0. The ranges decrease with increasing energy above 3 GeV. For both targets ^{28}Mg has a smaller F/B than ^{24}Na at the same energy, and has a larger range. The behavior of the ranges below 3 GeV is not clear, since there are data only at one energy; however, except for ^{24}Na from uranium, the range seems to be about the same at 0.7 GeV as at 3.0 GeV.

Comparing the recoil properties of the light fragments with those of the medium-mass nuclides one sees that there is a great similarity between the energy dependence of F/B for ^{131}Ba (and presumably other neutron-deficient nuclides) and that for ^{24}Na and ^{28}Mg. In contrast is the essentially energy-independent behavior of the recoil properties of ^{140}Ba, which is typical of neutron-rich nuclides. The energy dependence of the ranges of light fragments is also similar to that of ^{131}Ba, although the decrease in range of the former is not as large as the latter. Although this similarity in recoil properties does not demonstrate that a light fragment is the complementary product of a neutron-deficient nuclide in a two-body breakup of an excited nucleus, it suggests that they are formed in similar kinds of events.

Beg and Porile[12] have explained the change in recoil properties of such neutron-deficient nuclides in terms of fragmentation. In this view the decrease in mean kinetic energy as measured by the range is correlated with the onset of fragmentation, since the Coulomb repulsion between such partners is smaller than for binary fission. Moreover, the predominantly forward emission of the fragments also tends to lower the mean kinetic energy of the massive partner because of the resulting backward recoil. The decrease in the F/B ratio of the massive partner is also due to the backward recoil from the fragment emission. This explanation would require that the F/B ratio of the light fragment remain large at high bombarding energies. Instead we see that this ratio decreases in exactly the same way as it does for the heavy neutron-deficient nuclides.

Thus it does not appear to be the case that the decreased forward peaking of the neutron-deficient nuclides is *caused* by the emission of light fragments. Rather it seems more likely that both types of products are being affected similarly by some change in the nature of the reaction mechanism, which is such as to cause decreased momentum transfer to the struck nucleus as the incident energy increases. Recent measurements of the recoil properties of a number of nuclides from uranium[20,21] and of ^{22}Na and ^{24}Na from aluminum[24] also indicate that F/B decreases when the incident proton energy increases from 11.5 GeV to 300 GeV. It thus may be the case that such a decrease in forward momentum transfer is a general phenomenon connected with the approach to ultra-relativistic proton-nucleus collisions. This energy

region appears to have been reached at 28 GeV for the reactions studied here, based on the F/B values measured at that energy (Fig. 4).

Further work on the energy dependence of recoil properties is clearly needed before definite conclusions can be drawn. In particular, it would be desirable to obtain angular distributions and energy spectra using thin targets in order to learn more about how the intranuclear cascade may change with increasing incident energy.

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