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Charge Distribution and Recoil Properties in the Fission of ^{238}U
by 11.5 GeV Protons*

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Abstract - Cross-sections and thick-target recoil properties have been determined for a number of products in the mass region 100-115 from the interaction of ^{238}U with 11.5 GeV protons. The recoil ranges of the neutron deficient products are nearly a factor of two smaller than those of the neutron excessive products, suggesting a difference in reaction mechanisms. On the basis of this finding a charge dispersion curve consisting of two separate gaussians, peaking at $N/Z = 1.22$ and 1.40 , was constructed to fit the measured cross sections. The curve associated with the neutron excessive products is very similar to that obtained in the fission of ^{238}U by 450 MeV protons except for a reduction in the fission cross section by a factor of 2.4. The values of the average deposition energy, average total kinetic energy of the primary fragments, and average separation distance at scission obtained for these fragments from their recoil properties are nearly equal to the corresponding values at 450 MeV. Integration of the charge dispersion curve for the neutron deficient products indicates a 15% contribution to the total isobaric cross section at $A = 111$. The width of this distribution is less than half that of the curve for neutron excessive products. Many of the properties of the neutron deficient products are consistent with either spallation or fission of a nucleus in the heavy rare earth region but neither mechanism can explain all the results.

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I. Introduction

The distribution of nuclear charge among the products of the reactions of heavy elements with GeV protons has been investigated in recent years (1-7). The charge dispersion determined for products with $125 < A < 140$ formed from uranium exhibits two distinct peaks of approximately equal height on either side of the beta stability line (1,2). This finding contrasts with the results below about 0.7 GeV (1,8,9) where the charge dispersion is single-peaked.

The differentiation of the charge dispersion at $A \approx 130$ into twin peaks is considered to arise from at least two different processes, one of which is the normal fission process found in the hundreds of MeV range of bombarding energy. Recoil data for a number of fission products support this hypothesis (2,10-12). The ranges of various neutron deficient nuclides are found to be approximately a factor of two less than those of comparable neutron excessive products. Such differences are not observed in the hundreds of MeV range of bombarding energy, where the fission mechanism adequately accounts for the recoil properties of both classes of products (10,12-15). The recoil properties of neutron excessive products from GeV bombardments are likewise accounted for by the same fission mechanism (11,12). On the other hand, the mechanisms giving rise to the neutron deficient products at GeV energies are not as yet completely understood. Evidence has been cited for contributions from fission (1,11), spallation (6,11), and fragmentation (2).

The present study is concerned with the charge dispersion and average recoil properties of products in the mass region $100 < A < 115$ from the interaction of 11.5 GeV protons with uranium. The purpose was to determine the extent to which the findings in the $A \approx 130$ mass region also apply at lower mass numbers.

Previous charge dispersion measurements (5) in this mass region were consistent with either a double- or single-peaked curve. We have earlier reported the results of similar measurements at 450 MeV (9,13) and so can also comment on the energy dependence of the measured properties.

II. Experimental Procedure

The irradiations were performed in the internal beam of the ZGS at Argonne National Laboratory. The length of the irradiation period varied from 5 to 15 min. Bombardments were performed employing the "flin-flop mode" or a shadow block to prevent low energy protons from striking the target. The target assembly consisted of 0.001 inch thick depleted ^{238}U foils, surrounded by 0.001 inch aluminum catcher foils of high purity, followed by 0.001 inch monitor foils and wrapped with 0.001 inch aluminum.

The majority of the cross sections were determined relative to the ^{111}Ag cross section in recoil experiments in which the target was positioned perpendicular to the beam. In these experiments the catcher foils overlapped the slightly smaller target foil to insure complete capture of all recoils from the target. In the determination of the absolute cross section of ^{111}Ag , the target, catcher, and monitor foils were properly aligned before wrapping the target assembly. This cross section was determined relative to that of the $^{27}\text{Al}(\text{p},3\text{pn})$ monitor reaction whose value at 12 GeV is 8.6 mb (16). The activity of the monitor foils was determined by means of methane-flow proportional counters which had been calibrated for ^{24}Na by beta-gamma coincidence techniques (9). The ^{24}Na activity of the individual monitor foils in a given target assembly agreed to within two per cent indicating a reasonable alignment of the foils.

Activity measurements on the radiochemically separated samples were made with proportional counters in the case of the negatron emitters, ^{109}Pd , ^{111}Ag , ^{112}Ag , ^{113}Ag , ^{115g}Cd , and ^{115m}Cd , with a 3 in. x 3 in. NaI (Tl) detector connected to a 400 channel analyzer in the case of ^{101}Pd , ^{112}Pd , ^{111}In , ^{107}Cd and ^{115g}Cd ; and with an x-ray detection system consisting of a thin sodium iodide crystal connected to a single channel analyzer in the case of ^{100}Pd , ^{103}Pd , and ^{105}Ag . The details about the radiochemical separations employed and the calibration of the various detectors have been described elsewhere (9). The decay properties of the observed nuclides are summarized in Table I.

III Results

A. Cross Sections

The activities of each product were corrected for chemical yield, detector efficiency, recoil loss, aliquot, absorber (^{105}Ag , ^{109}Pd , ^{107}Cd , ^{115m}Cd , ^{115g}Cd), parent-daughter factors (^{101}Pd , ^{103}Ag , ^{107}In , ^{112}Pd), and disintegration conversion factors listed in Table I, and the cross sections calculated by the standard relationships. In the case of ^{111g}Pd and ^{111m}Pd , two simultaneous equations (9) relating the observed ^{111}Ag activities to the initial activities at the end of bombardment of these nuclides were solved. The activities of ^{101}Pd , ^{103}Ag , and ^{107}In at the end of bombardment were obtained by correcting the values obtained from their respective daughter activities for decay up to the time of separation.

The results are presented in Table II along with previous determinations at GeV energies (5,17,18). The measured cross sections are either independent (i) or cumulative (c). In several cases there may be some ambiguity as to the

mode of formation. These cases have been identified in the table. The cross section of ^{111m}Pd is considered to be independent since the decay of ^{111}Rh is predominantly (99%) to the ground state of ^{111}Pd .

The quoted errors are standard deviations based upon the number of separate determinations listed in Table II. In the case of ^{103}Ag , the quoted error is the standard deviation of duplicate determinations from the same target. The overall uncertainty in the cross section measurements is estimated to be 10 per cent for the neutron emitters and ^{111}In and 20 per cent for the other nuclides. This is comprised mainly of the uncertainties in the efficiencies of the detectors (5 per cent); an uncertainty in the cross section of the monitor reaction (5 per cent) and the uncertainty in the conversion of the observed activity to disintegration rate for nuclides detected via x-rays or gamma rays (10 per cent).

The present data are in reasonably good agreement with previously reported data within the overall uncertainty of the measurements.

B. Recoil Properties

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 quantities $2W(F+B)$ and F/B , listed in Table III. The target thickness is $W \text{ mg/cm}^2$. The quoted values have been corrected for scattering (14) (4%) and edge (19) (0.5%) effects.

The analysis of the recoil data is based on a previously developed (20, 21, 14) velocity vector model embodying the two-step mechanism usually invoked in high-energy fission. Equations have been derived on the basis of this model relating

the measured recoil properties to the mean range of the fragment in the moving frame, R , and to the velocity ratio, $\eta_{\parallel}$. This quantity is defined as $\eta_{\parallel} = v_{\parallel}/V$ where $v_{\parallel}$ is the component of the average velocity of the cascade product along the beam direction and V is the average velocity of the fragment in the moving frame. These two recoil parameters have been evaluated using exactly the same procedure described in detail in an earlier report (13) and are summarized in Table III. Recent (11) differential range measurements indicate that at GeV energies the velocity distributions of neutron deficient products in the fission region are very broad. The effect of such broad distributions on the vector model analysis of thick target recoil data has been explored (22). It is expected on the basis of this analysis that the vector-model formulation may overestimate the values of R and V by less than 5%. On the other hand, the values of $v_{\parallel}$ may be underestimated by as much as 20%.

IV. Discussion

A. Analysis of Cross Section Data

1. Charge Distribution of Normal Fission Products

Since the pattern of recoil ranges at GeV energies suggests the occurrence of two distinct mechanisms it is reasonable to interpret the measured cross sections from the same point of view. Each mechanism will result in a characteristic charge dispersion curve and the experimentally observed curve will be their weighted sum.

The recoil ranges of the negatron emitters are of comparable magnitude as those of fission products formed at 450 MeV. It is therefore assumed that the cumulative and independent cross sections of these products are consistent with the same type of charge dispersion curve as was found (9) to fit the

results at 450 MeV.

The cross sections of the neutron emitters for mass chains 111 and 112 were thus fitted to the gaussian expression

$$\sigma(Z) = \sigma_{\max} \exp - [(Z - Z_p)^2 / 2S^2] \quad (1)$$

where $\sigma(Z)$ is the independent cross section of the product of nuclear charge Z ; $\sigma_{\max}$ is the cross section of the product with the most probable charge Z_p and S is the standard deviation of the distribution. The solution of Eq. (1) requires at least three yield measurements for a given mass chain. Since only two yield measurements were available for each mass chain, S was assumed equal to 1.233, the value found for mass chain 111 in the 450 MeV proton fission of uranium (9). As noted in this study, S appears to be independent of bombarding energy, or more precisely, of the spectrum of excitation energies generated by the initial interaction. Since, as shown in Section IV B2, the average deposition energies of cascade nuclei leading to these products are only slightly higher than the corresponding values with 450 MeV protons, (13,14) the assumption should not be in serious error. The value of S corresponds to a full width at half-maximum, (FWHM), of the charge distribution of 2.90 Z units.

Equation (1) was solved by means of successive approximations such that the calculated independent yields of a product and its progenitors add up to the cumulative yield of the observed product. The results are presented in Table IV in which column 1 gives the number of the mass chain; column 2, the value of $\sigma_{\max}$; column 3, the total isobaric yield expected for "normal fission" products; column 4, the calculated value of Z_p ; and column 5, the neutron-to-proton ratio, N/Z_p , of the most probable product. The uncertainty in the Z_p values

is approximately 0.5% of the quoted values, corresponding to the limits which reproduce the experimental data within their errors. The Z_p values for mass chain 109 and 115 were calculated assuming a FWHM of 2.90 Z units and a total isobaric chain yield of 29.4 mb, the average value found for mass chains 111 and 112. Since the cumulative yield of ^{113}Ag is expected to be low because of some direct decay of $^{113\text{m}}\text{Ag}$ to ^{113}Cd , the Z_p value for this mass chain was not calculated.

The N/Z_p values for the four mass chains are relatively constant at 1.402 ± 0.008 . The independent cross section in this mass region of a product of mass A and charge Z, $\sigma_{(A,Z)}$, may therefore be expressed by the equation

$$\sigma_{(A,Z)} = (9.5 \pm 0.3) \exp - [(A/2.40 Z)^2 / 17.80] \quad (2)$$

obtained by substituting $A/2.402$ for Z_p in Eq. (1). Equation (2) implies a gaussian distribution of not only isobaric yields but isotopic yields as well. This result is a consequence of the constant isobaric yield and N/Z_p ratio in this mass region.

The calculated cross sections of the negatron emitters are compared with the corresponding experimental values in Fig. 1. The curve centered about $N/Z = 1.40$ represents the isobaric yield distribution of "normal fission" products calculated by means of Eq. (2) for $A = 111$. The experimental data for mass chains 109, 111, 112, 113 and 115 are plotted in the same figure for an approximate comparison since in such a representation the wings of the independent yield curve vary slightly for each mass chain. With the exception of the cross section for ^{113}Ag , the experimental value of which is expected to be low as discussed above, excellent agreement is found between the calculated and experimental values within their listed uncertainties, 3 to 4 per cent.

2. Charge Distribution of Neutron Deficient Nuclides

Since there were not more than two isobaric yields determined for the neutron deficient nuclides for any mass chain investigated and since no data are available on the charge dispersion width to be expected for these products, a somewhat different procedure was followed. The isotopic yield distribution was assumed to be gaussian and the three neutron deficient palladium yields were fitted to the expression:

$$\sigma(A) = \sigma_{\max} \exp - [(A - A_p)^2 / 2s^2] \quad (s)$$

where $\sigma(A)$ is the independent cross section of a product of mass number A and $\sigma_{\max}$ is now the cross section of the most probable product A_p . The "normal fission" contribution calculated by means of Eq. (2) was first subtracted from the cumulative yields of these products. This contribution amounted to less than 10%. Equation (3) was then solved by successive approximations until the calculated independent yields of the observed product and its progenitors summed up to the experimental cumulative cross section. A similar procedure was followed for the neutron deficient silver products. The results are summarized in Table IV. Since $\sigma_{\max}$ and N_p/Z are relatively constant for these two isotopic charge chains, Eq. (3) may be generalized to give:

$$\sigma(A, Z) = (3.61 \pm 0.003) \exp - [(A - 2.221 Z)^2 / 3.040] \quad (4)$$

Equation (4), like Eq. (2), implies a gaussian distribution of both isobaric and isotopic yields.

A comparison between the cross sections of the neutron deficient products calculated from Eq. (4) and experimental data is made in Fig. 1. The curve centered about $N/Z = 1.22$ represents the charge distribution of neutron deficient products calculated for $A = 111$. The experimental cross sections of

all the neutron deficient products are plotted in the same figure for an approximate comparison. Equation (4), derived from the data of silver and palladium products, is found to adequately account for the cross sections of ^{107}In , ^{107}Cd , and ^{111}In , after adding relatively small contributions from the normal fission process.

The reported cross sections (5) of ^{109}In and ^{109}Cd , and in particular their ratio, are found to be significantly different from those expected from the proposed charge distribution. A redetermination of these values as well as the measurement of the cumulative yield of ^{111}Sn is suggested. The latter cross section would permit an evaluation of the independent yield of ^{111}In which along with the independent yield of ^{109}Cd should establish whether a minimum in the charge distribution occurs at $N/Z = 1.27$, as predicted by the present analysis. In the absence of more detailed results the decomposition of the charge dispersion into two distinct curves must be considered tentative. The cross sections determined in the present work could also be adequately accounted for by a single broad charge dispersion curve.

3. Charge Dispersion for $A=111$

The overall charge dispersion in the mass region $A=111$ is obtained as the sum of the two separate gaussians and, as shown in Fig. 1, exhibits two peaks. The following comments may be made about this curve:

The total isobaric yield at $A = 111$ is estimated as 33.5 ± 3.4 mb. This is comprised of a 28.5 mb contribution associated with the normal fission process and a 5.0 mb yield of the neutron deficient peak. We conclude that approximately 85% of the products in this mass region are produced by the normal fission process.

The full-width at half maximum of the neutron deficient peak is 1.31 Z units. This is over a factor of two narrower than the value of 2.90 Z units derived for the normal fission peak.

These results may be related to other high-energy data. The isobaric yield at $A = 109$ has been previously determined for 3 and 28 GeV protons (5). Cross sections of respectively 32.2 ± 3.8 mb and 27.2 ± 3.5 mb were reported at these energies. The present isobaric cross section at essentially the same mass number is somewhat higher but agrees within the limits of error.

Crespo et al (11) have recently measured the differential range of ^{103}Pd from the interaction of uranium with 2.2 GeV protons. These workers decomposed their range curve into contributions from fission and spallation and on this basis estimated a 2% contribution from the latter to the total isobaric yield at $A = 103$. While this is a substantially lower estimate than that obtained in the present work, it is reasonable to expect that the contribution from spallation will increase between 2 and 12 GeV.

The present results may be compared with similar data (1,2) for the mass region $125 < A < 140$. While the charge dispersion curve exhibits double maxima in both regions the magnitude of the neutron deficient peak is comparable to that of the neutron excessive peak at the higher mass numbers. In addition, the widths of the two peaks are comparable at $A \approx 130$, in contrast to the situation at $A \approx 111$. These results are consistent with a spallation mechanism for the formation of the neutron deficient products since the importance of this process should increase with decreasing mass difference between target and products.

The charge distribution is compared with that previously determined (9) at

450 MeV in Fig. 2. The striking change from a single-peaked to a double-peaked curve is evident. The total isobaric yield associated with the neutron-excessive peak decreases by a factor of 2.3 between 0.45 and 11.5 GeV. This may be compared with a value of 1.7 for the corresponding decrease in the total binary fission cross section between these same energies (23). It may also be noted that the value of Z_p associated with the normal fission peak is rather insensitive to the proton energy, decreasing from 1.42 to 1.40 over the energy range of interest. This small variation corresponds to an increase in Z_p of about 0.15 Z units which is consistent with the near constancy of the deposition energies, discussed in section IV B2.

Since spallation is believed to contribute to the yield of the neutron deficient products it is of interest to compare the charge dispersion associated with these products with that for a known spallation process. The proposed charge distribution, plotted as an isotopic distribution of the neutron deficient palladium nuclides is compared in Fig. 3 with a similar curve obtained for the interaction of silver with 400 MeV protons (24). The curves correspond to gaussian distributions fitted to the experimental data, denoted by the circles and squares. The rise in the charge distribution from ^{238}U at $N/Z > 1.28$ is due to the increasing contribution from normal fission. It is seen that the two curves are quite similar, peaking at $N/Z \sim 1.22$ and having a width of about 2.90 amu. A similar result was obtained by Rudstam and Sorensen (6) from an isotopic yield study of neutron deficient iodine products from the bombardment of uranium, lanthanum and cesium with 18 GeV protons. The authors concluded that the neutron deficient products were produced from uranium by a spallation mechanism involving long evaporation chains.

Another possible mechanism for the formation of the neutron deficient

nuclides is normal fission of the type obtained at 450 MeV but from a much lighter fissioning nucleus than that leading to the neutron excessive products. Such a fissioning nucleus could be produced by a long cascade-evaporation chain accompanied, perhaps, by the prompt emission of light fragments. The charge dispersion curve expected for such a mechanism may be estimated by extrapolating the results previously obtained at 450 MeV for (9) ^{238}U and (15) ^{208}Pb to lower masses. A comparison of these two curves reveals a trend of decreasing N/Z_p and FWHM with decreasing target mass. The curve obtained for the isotopic yields of palladium nuclides for a fissioning nucleus with $A = 170$ is shown in Fig. 3. The curve peaks at a slightly larger N/Z value and is somewhat narrower than the experimental curve. In view of the highly approximate nature of this extrapolation the resulting curve does not appear inconsistent with the present results.

B. Analysis of Recoil Data

1. Dependence of $2W(F+B)$ on N/Z of Product

The dependence of the recoil range on the composition of the product may be examined in light of the charge dispersion analysis. The data in Table III indicate that there is nearly a factor-of-two difference between the ranges of the neutron excessive and deficient products and that within each group the ranges are practically constant. The average value of $2W(F+B)$ for products from the normal fission distribution was calculated to be $8.9 \pm 0.2 \text{ mg/cm}^2$. A value of $4.8 \pm 0.5 \text{ mg/cm}^2$ was estimated for products from the other distribution and represents the average $2W(F+B)$ of neutron deficient products after correction for contribution from the normal fission process.

The dependence of the range on N/Z was calculated by weighting these average values by the relative contribution from each charge dispersion. The results are shown as the solid curve in Fig. 4. The experimental values are plotted at

the average N/Z of the products, as determined from the charge dispersion.

The step-function dependence of $2W(F+B)$ on N/Z of the product predicted by the present analysis implies that, in the mass region $100 < A < 115$, the sudden drop in range should occur over 2 Z units (e.g. ^{111}Cd should have a normal fission range whereas the range of ^{111}Sn and higher Z isobars should be lower by a factor of two). A similar drop is estimated to occur in isotopic range studies over 2 mass units, (e.g. $^{112}\text{In} - ^{110}\text{In}$). Such a step-function dependence of the range on mass number has been observed by Brandt in a study of the recoil ranges of iodine isotopes from 18 GeV proton bombardments of uranium (10). An abrupt change in the range was found to occur over 2 mass units, between ^{124}I and ^{122}I . Similar results have also been reported recently for the antimony isotopes formed in the interaction of uranium with 18 GeV protons (12). The abruptness of the transition between the two types of ranges depends, of course, on the relative magnitude and extent of overlap between the two distributions.

Also shown in Fig. 4 are the ranges previously measured at 450 MeV for a number of the same products (13). The previously noted decrease (2,10,13) by a factor of approximately 2 in the ranges of neutron deficient products between 0.5 and 12 GeV is apparent for ^{103}Pd and ^{111}In . By contrast, the ranges of the neutron excessive products decrease by at most a few percent over the same energy interval.

2. Kinetic and Deposition Energies

The ranges in Table III were converted to kinetic energies, T , by means of a range-energy relation derived from thermal neutron data. The procedure has been described in detail in a previous publication (13) and includes a correction for the difference between the average charges in thermal and high-energy fission. The application of this relation to the neutron deficient

products is somewhat questionable and the results should be regarded as approximate. The kinetic energies are summarized in Table V where the results for the neutron deficient products have been corrected for a small contribution from normal fission.

A simple calculation has been described (14,13) whereby the kinetic energies of the observed products are used to derive the average total kinetic energy of the primary fragments, $\langle T^{\text{tot}}(Z,A') \rangle$ and their mean separation distance at scission, D . The calculation of the first quantity is based on the assumption of a particular average fissioning nucleus, $^{234.4}\text{U}$, an estimation of the mass of the primary fragment corresponding to a particular product based on the excitation energy associated with its formation, and conservation of momentum considerations. The quantity D is then determined from a straightforward application of Coulomb's law. In view of the close similarity between the properties of the neutron excessive products at 1155 and 0445 GeV, this formalism was applied to the present results using the same procedure and parameters as at 450 MeV (13,14).

The results for the neutron excessive products are summarized in Table V. If these results are weighted by their fission yield, average values for A_{fission} may be obtained. The average total kinetic energy release is 156 ± 3 MeV with an absolute uncertainty of 8%. The corresponding average value of D is 19.4 ± 0.6 fermi. Within the experimental errors, the values are equal to the comparable values, 160 ± 2 MeV, and 19.1 ± 0.1 fermi found at 450 MeV (13,14).

The average deposition energy, E^* , associated with the products of interest may be obtained from the relation

$$E^* = 0.85 E_p p_p / P_p \quad (5)$$

where E_p and P_p are respectively the kinetic energy and momentum of the incident proton and $p_{//}$ is the forward component of momentum of the cascade product.

The above relation was derived by Porile (25) on the basis of Monte Carlo cascade calculations (26) performed up to an energy of 1.8 GeV. The validity of this relationship at 12 GeV has not been demonstrated and the results must be regarded in light of this fact. The average values of $v_{//}$ are given by $v_{//} = \eta V$ with the V values obtained from the calculated kinetic energies. The E^* values were then obtained from Eq. 5, using a mass of 236 amu to convert $v_{//}$ to $p_{//}$, and are summarized in Table V. The quoted errors reflect the experimental uncertainties in $2W(F+B)$ and F/B and do not include whatever error is inherent in the use of Eq. (5). In addition, the values of E^* associated with the formation of the neutron deficient products may be too low by some 20% because of the broad distribution of ranges (22).

The E^* values may be compared with similar results obtained at 450 MeV (13,14) and the latter are also summarized in Table V. The deposition energies associated with the formation of the neutron excessive products show only a slight increase with bombarding energy. This can be seen somewhat more clearly by computing the average deposition energy associated with the formation of the most probable product at a given mass number, $E^*(Z_n, A)$. This quantity has been carefully determined for the mass region of present interest at 450 MeV from recoil measurements on many products not included in Table V (14). A similar determination may be made at 12 GeV following the procedure outlined elsewhere (13,14). It is found that the weighted average of $E^*(Z_n, A)$ for $100 < A < 115$ is 146 ± 13 MeV. This is only slightly larger than the value, 120 ± 3 MeV, found at 450 MeV (14), suggesting that the selectivity of the fission process

for cascade events leading to the most probable product in GeV bombardments is quite similar to that found in the hundreds of MeV range of bombarding energy.

The E^* values associated with the neutron deficient products are rather perplexing. On the average, these values are not any larger than those for the neutron excessive products. Also, the values decrease as the bombarding energy increases from 0.45 to 11.5 GeV. This surprising result persists even if a 20% allowance is made for the effect (22) of the broad range distributions observed for these products (11) at GeV energies. If the neutron deficient products are formed either by spallation or fission of a low Z element then one might expect E^* values above 0.4 GeV. This large discrepancy may reflect a change in the relation between $v_{||}$ and E^* from that found below 2 GeV or perhaps even the failure of the two-step model commonly invoked in high energy spallation and fission. Indeed, Alexander et al (2) have cited this type of result as evidence for a fragmentation process. The preferential forward emission of light fragments during the initial interaction leads to a concomitant reduction in the $v_{||}$ values of the heavy product and thus to an underestimation of E^* .

The results of other recoil measurements of neutron deficient products may be examined in the above terms. The data of Brandt (10) on iodine isotopes produced in 18 and 0.6 GeV proton irradiations of uranium may be analyzed to yield E^* values which show a pattern that is similar to that reported in the present work. The results of Alexander et al (2) for iodine isotopes formed at 6 GeV tend to exhibit the presently observed trend although in much less severe fashion. On the other hand, the results of Hagebo and Ravn (12) on antimony isotopes at 18 and 0.6 GeV exhibit the expected increase if E^* with bombarding energy. Their results at 18 GeV are also similar to the lower energy results in that E^* varies

inversely with product N/Z . The differential range measurements at 2.2 GeV by Crespo et al. (11) also yield this inverse dependence. The differences between these studies appear to lie in the measured F/B values. In those studies where low E^* values are reported for the neutron deficient products F/P ratios are close to unity while higher E^* values result from F/P greater than about 1.3. All the above studies agree on the low ranges of the neutron deficient products. It would thus appear that further experimental work may be required to settle this point.

3. Discussion of the Recoil Properties of Neutron Deficient Products

The charge distribution of the neutron deficient products has been found to be consistent with two different processes: spallation and fission of a low- Z nuclide. The prompt emission of light fragments may, in addition, occur during the initial stage of either mechanism. In this section we investigate the recoil properties expected from these processes.

The theory of nuclear evaporation has been quantitatively formulated and a calculation of the recoil ranges of spallation products has been developed (27,28). In the absence of Monte Carlo cascade calculations at 12 GeV, a highly approximate procedure was adopted to simulate the average results of the cascade. The distribution of cascade products was replaced by a single average residual nucleus. Its identity was determined by extrapolation of the curve relating the average number of nucleons emitted in the cascade to the average excitation energy of the residual nucleus determined by Metropolis et al (26) for energies up to 1.8 GeV. The value of E^* was chosen to optimize the yield of products in the mass range of interest. On the basis of these considerations ^{203}Pb with $E^* = 1.8$ GeV was adopted as the average cascade progenitor of products

with $A = 100-115$ from the interaction of ^{238}U with 12 GeV protons.

The velocity components of the residual nucleus were obtained from the previously derived (25) $v_{\perp} - E^*$ relation, as summarized by Eq. 5. The transverse component of velocity of the residual nucleus, $v_{\perp}$ was obtained from the predicted variation of $v_{\perp}/v_{\parallel}$ with E^* (25). Once again, it should be emphasized that these relations have been derived from cascade calculations (26) performed for 0.4 - 1.8 GeV protons. The adopted values are summarized in Table VI.

The residual nucleus resulting from the cascade deexcites by the evaporation process. The distribution of products and concomitant recoil ranges resulting from this process was determined by a Monte Carlo calculation based on the code by Dostrovsky et al (29) as modified by Porile (27,28) to keep track of the velocity of the evaporating nuclide. The calculation considered the emission of neutrons, protons, deuterons, tritons, ^3He , ^4He , and ^7Li fragments at each step of the deexcitation process. The emission probability of ^7Li was increased to include the emission of all bound nuclides with $A \leq 10$. This was accomplished by increasing the statistical weight of this fragment by a factor of 5, as discussed in detail elsewhere (27,30). The evaluation of the emission widths of individual heavy fragments is impractical in terms of the required computer time.

At the end of the deexcitation process the kinetic energy and direction of motion of the product with respect to the beam axis were computed. The kinetic energy was converted to the range in uranium by means of a range-energy relation based on the calculation of Lindhard et al (31) for low-energy products and on the above mentioned relation for fission fragments for high energy products. The ranges were then projected along the beam direction according to a procedure discussed elsewhere (32,33) and the results for 1000 iterations were averaged

to yield average projected ranges, FV and BW , for comparison with experiment. In performing this comparison both the calculated and experimental values of FV and BW were averaged over all the appropriate spallation products. The calculated values were thus obtained by averaging the results for all products with $A = 100-115$ while the experimental values are an average for all the neutron deficient products, corrected wherever necessary for a contribution from fission.

The results are summarized in the first line of Table VI. It is seen that the calculation overestimates the value of FV and grossly underestimates that of BW . The calculation can be brought into somewhat better agreement with experiment by reducing the values of v_{α} and v_{β} and maintaining the excitation energy at 1.8 GeV. It is possible in this fashion, as shown in the table, to bring the calculated value of FV into perfect agreement with experiment. However, the predicted value of BW is still too small by nearly a factor of six. In order to eliminate this large forward backward asymmetry without a substantial reduction in the magnitude of the projected ranges, the deexcitation process would have to include the abundant emission of much heavier fragments than those included in the calculation.

The ranges expected from fission of a low- Z nuclide may be estimated by examination of the dependence of the product range on target Z obtained at 450 MeV (19). The experimental values of $2W(F+B)$ are consistent with a fissioning nucleus in the Z range 65-70, in approximate agreement with the estimate based on the charge distribution. This finding is consistent with the work of Crespo et al (11) who account for the mean velocities of neutron deficient products obtained from differential range measurements by means of a similar mechanism.

Although this mechanism satisfactorily accounts for the measured ranges it requires F/B values that are much larger than the experimental ones, although not as large as those expected from spallation. Once again, lower F/B values would be predicted if the $v_{//} - E^*$ relation obtained below 2 GeV does not apply at 12 GeV or if light fragments are emitted preferentially in the forward direction during the initial interaction.

Another difficulty inherent in the formation of both neutron excessive and deficient nuclides by fission is that this mechanism implies a continuous distribution of fissioning nuclei, ranging from nuclides close to the target to those perhaps 70 mass units removed from it. This would in turn imply a continuous increase of range with the N/Z of the product instead of the observed step function. One would also expect a single, broad, charge dispersion curve instead of two distinct peaks. It may of course turn out, due to the various factors determining spallation-fission competition, that in the interaction of ^{238}U with 12 GeV protons fission is restricted to nuclei with $Z \sim 92$ or $Z \sim 65-70$. However, we have no evidence supporting this hypothesis.

V. Conclusions

The charge dispersion in the mass region $100 < A < 115$ from 11.5 GeV proton bombardments of ^{238}U is consistent with a double-peaked curve similar to that reported for the mass region $A \sim 130$. The height of the neutron excessive peak is estimated to be approximately 2.6 times greater than that of the neutron deficient peak and its width, approximately a factor of 2 greater. The corresponding most probable N/Z for the two peaks are 1.22 and 1.40. The latter value is comparable to the value 1.42, found for this mass region in 450 MeV bombardments (9). The decrease in N/Z_p at 11.5 GeV amounts to an increase in Z_p of 0.5 Z

units for normal fission products.

The total isobaric yield of mass chain 111 is measured to be 33.5 ± 3.4 mb. This comprises 28.5 mb (85%) attributable to the normal fission process and 5.0 mb (~15%) attributable to other processes. The fission cross section about symmetric fission at 11.5 GeV is a factor of 2.4 lower than the value at 450 MeV (9), which is comparable to the reduction in the total fission cross section (23).

The recoil ranges of neutron deficient products are found to be approximately a factor of 2 less than at 450 MeV (13) and than of neutron excessive products at 12 GeV whereas the ranges of the latter are only slightly smaller than at 450 MeV (13). On the basis of the resolution of the charge dispersion into two component distributions, with $2W(F+B)$ values differing by a factor of nearly two, a step function dependence of range on N/Z of the product or on Z and A of an isobaric mass or isotopic charge chain is predicted. This sudden change in range is expected to occur over two mass or two charge units, as found experimentally.

From an analysis of thick-target recoil properties, the following quantities have been determined for the neutron excessive products: the average deposition energy of cascade nuclei leading to the formation of the given product and to that of the most probable product for a given mass chain, the average total kinetic energy release, and the separation distances of the primary charge centers. The values of E^* (Z_0, A) calculated from the observed values of E^* are relatively constant at 146 ± 13 MeV. This value is only slightly larger than the value (13,14) 120 ± 3 MeV determined at 450 MeV. The average values of the total kinetic energy of the primaries, 156 ± 3 MeV, and their separation distance at scission, 19.4 ± 0.6 fermi, are found to be equal, within the experimental errors, to the values (14) 160 ± 2 MeV and 19.1 ± 0.1 fermi determined at 450 MeV, providing further

evidence for the similarity of the process at the two bombarding energies.

The analysis of the results for the neutron deficient products leads to the following conclusions: 1. The charge dispersion obtained after subtraction of the contribution from normal fission closely resembles that obtained in spallation reactions. However, a rather similar curve is expected for fission of cascade evaporation products lying in the heavy rare-earth region. 2. The experimental value of F/B is consistent with that expected for spallation but the forward to-backward ratio is too low by a factor of ~ 6 . This discrepancy could be reduced if modifications in the calculation lead to the copious evaporation of fragments with $A > 10$. The measured values of $2^{1/2}(F+B)$ are also in line with the expectation for fission of heavy rare earth nuclei but once again, larger F/B values would be expected. 3. The deposition energies obtained for these products are far too low to be consistent with either of these two mechanisms. The difficulty is ascribed to the small values of F/B which may be indicative of a breakdown of the assumed $v_{rel} - E^*$ relation (25) or even of the two-step model of high-energy reactions.

If thus appears that the production of the neutron deficient products at high energies is not completely consistent with either spallation or fission of low Z nuclei. It is quite likely that both these processes make a contribution but additional mechanisms, such as fragmentation, may also be of importance. It is apparent that further work is needed to clarify this problem.

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Table I

Decay Properties of Observed Radionuclides^a

Nuclide	Mode of Decay	$T_{1/2}$	Radiation Measured	D/A ^b
¹⁰⁰ Pd	100% E.C.	3.63 d.	X-ray	0.801 ^c
¹⁰¹ Pd	96.8% E.C.; 3.2% β^+	8.4 h.	307 KeV γ (daughter)	1.20 ^d
¹⁰³ Pd	100% E.C.	17.0 h.	X-ray	1.23
¹⁰³ Ag		1.1 h.	daughter	
¹⁰⁵ Ag	100% E.C.	40.0 d.	X-ray	1.27
¹⁰⁷ Cd	99.7% E.C.; 0.3% β^+	6.7 h.	X-ray	1.02
¹⁰⁷ In		32 m.	daughter	
¹⁰⁹ Pd	100% β^-	13.5 h.	β^-	1.00
^{111g} Pd	100% β^-	22 m.	daughter	
^{111m} Pd	32% β^- , 68% IT	5.5 h.	daughter	
¹¹¹ Ag	100% β^-	7.5 d.	β^-	1.00
¹¹¹ In	100% E.C.	2.81 d.	247 KeV γ	1.06
¹¹² Pd	100% β^-	21.0 h.	618 KeV γ , (daughter)	
¹¹² Ag	100% β^-	3.2 h.	β^-	1.00
¹¹³ Ag	100% β^-	5.3 h.	β^-	1.00
^{115m} Cd	100% β^-	43.0 d.	β^-	1.00
^{115g} Cd	100% β^-	2.3 d.	β^- 582 KeV γ	1.00 2.15

^a Based on properties described in Nuclear Data Sheets, compiled by K. Wav et. al. (Printing and Publishing Office, National Academy of Sciences - National Research Council, Washington 25, D. C.).

^b Disintegration per emitted radiation. Values of nuclides measured via X ray emission contain corrections for percentage electron capture, fraction of K capture, fraction of K vacancies resulting from gamma rays of associated daughter, and fluorescence yield of the daughter nuclide.

^c Based on decay scheme proposed by J. S. Evans and P. A. Naumann, Phys. Rev. B138, 1017 (1965).

^d Based on decay scheme proposed by J. S. Evans et al., Phys. Rev. B138, 9 (1965) and J. S. Evans and R. A. Naumann, Phys. Rev. B140, 559 (1965).

Table II

Formation Cross Sections of Products from GeV Proton Bombardments of Uranium

Nuclide	Bombarding Energy (GeV)	3 ^a	5.7 ^b	11.5 ^c	28 ^a
¹⁰⁰ Pd (c)		0.88 ± 0.12		0.90 ± 0.02 (3)	1.4 ± 0.1
¹⁰¹ Pd (c)		2.0 ± 0.1		2.60 ± 0.05 (2)	2.7 ± 0.4
¹⁰³ Pd (c)		3.6 ± 0.2 (3.7) ^d		5.10 ± 0.02 (4)	4.1 ± 0.2 (4.2) ^a
¹⁰⁹ Pd (c)		21.4 ± 2.5	19.1	22.9 ± 0.7 (2)	16.8 ± 2.1
^{111m} Pd (i)		5.5 ± 0.3	2.2	5.6 ± 0.8 (2)	3.9 ± 0.2
^{111g} Pd (c)				11.3 ± 0.2 (3)	
¹¹² Pd (c)		16.9 ± 1.5	1.0	13.8 ± 0.1 (5)	14.3 ± 0.4
¹⁰³ Ag (c)				1.9 ± 0.1 (1)	
¹⁰⁵ Ag (c)				5.3 ± 0.15 (2)	
¹¹¹ Ag (c)			21.5 ± 3 ^e	24.3 ± 0.3 (3)	
¹¹² Ag (i)			7.9	9.35 ± 0.09 (5)	
¹¹³ Ag (c) ^f			12.4	13.8 ± 0.1 (5)	
¹⁰⁷ Cd (c)				5.3 ± 0.1 (2)	
^{115m} Cd (i) ^g		6.6 ± 0.7		6.4 ± 0.3 (2)	5.6 ± 0.1
^{115g} Cd (c)		14.2 ± 0.9		15.8 ± 0.2 (2)	13.3 ± 0.8
¹⁰⁷ In (c)				1.3 ± 0.3 (2)	
¹¹¹ In (c)		4.6 ± 0.1	4.9	5.84 ± 0.09 (4)	5.0 ± 0.5

^a Reference 5^b References 17 and 18. The reported cross sections were adjusted to the monitor cross section used in the present study.^c Present study. (Values in parenthesis refer to number of determinations).^d Value corrected for radiation abundance used in present study.^e Reference 2^f Partial cumulative yield.^g Partial independent yield. (It is estimated that there is an 8% contribution of the cumulative parent yield to the observed cross section).

Table III

Recoil Parameters of Uranium Fission Products

Recoil properties (corrected)			Recoil parameters	
Nuclide	$2W(F + B)$ (mg/cm ² U)	F/B	R (mg/cm ² U)	$\eta_{//}$
Neutron Excessive Products				
¹⁰⁹ Pd	8.71 ± 0.12 (2) ^a	1.10 ± 0.02	8.67 ± 0.12	0.022 ± 0.004
¹¹² Pd	8.98 ± 0.06 (4)	1.07 ± 0.01	8.96 ± 0.06	0.015 ± 0.002
¹¹¹ Ag	8.75 ± 0.03 (2)	1.105 ± 0.005	8.70 ± 0.03	0.0226 ± 0.0002
¹¹² Ag	9.21 ± 0.19 (2)	1.10 ± 0.02	9.16 ± 0.19	0.022 ± 0.004
¹¹³ Ag	8.87 ± 0.15 (2)	1.08 ± 0.02	8.84 ± 0.15	0.017 ± 0.004
Neutron Deficient Products				
¹⁰⁰ Pd	4.52 ± 0.05 (2)	1.12 ± 0.03	4.50 ± 0.05	0.025 ± 0.004
¹⁰¹ Pd	5.46 ± 0.06 (2)	1.14 ± 0.02	5.43 ± 0.06	0.029 ± 0.004
¹⁰³ Pd	5.22 ± 0.06 (2)	1.130 ± 0.003	5.20 ± 0.06	0.028 ± 0.001
¹⁰³ Ag	4.53 ± 0.11 (1)	1.13 ± 0.02	4.50 ± 0.11	0.028 ± 0.003
¹⁰⁵ Ag	5.64 ± 0.06 (2)	1.13 ± 0.03	5.62 ± 0.06	0.028 ± 0.005
¹¹¹ In	4.84 ± 0.01 (2)	1.155 ± 0.002	4.82 ± 0.01	0.033 ± 0.001

^a Number of separate determinations

Table IV
Properties of Isobaric Mass and Isotopic Charge Chains from
11.5 GeV Proton Bombardments of ^{238}U

Neutron Excessive Products					
A	σ_{max} (mb)	σ_{tot} (mb)	Z_p	N/Z_p	
109		(29.4) ^a	45.58	1.391	
111	9.2	28.5	46.24	1.401	
112	9.8	30.2	46.63	1.402	
115		(29.4)	47.65	1.413	
Neutron Deficient Products					
Z	σ_{max} (mb)	σ_{tot} (mb)	FWHM (Mass Units)	A_p	N_p/Z
46	3.58	11.1	2.90	102.09	1.219
47	3.64	11.3	(2.90)	104.49	1.223

^a Values in parenthesis are assumed

Table V

Average Kinetic Energy and Cascade Deposition Energy for
Formation of Fission Products

Nuclide Isolated	T_{corr} (MeV)	$\langle T^{\text{tot}}(Z, A') \rangle$ (MeV)	D (fermi)	v_{c} (MeV/amu) ^{1/2}	E^* (MeV)	E^{*a} (MeV)
Neutron Excessive						
¹⁰⁹ Pd	71.0	151.4	20.1	0.025 ± 0.05	140 ± 27	118 ± 2
¹¹² Pd	73.8	155.6	19.6	0.018 ± 0.02	100 ± 14	99 ± 3
¹¹¹ Ag	71.5	155.5	19.6	0.026 ± 0.00	146 ± 4	116 ± 11
¹¹² Ag	78.2	167.6	18.2	0.026 ± 0.05	145 ± 28	
¹¹³ Ag	73.1	157.3	19.4	0.020 ± 0.05	113 ± 27	
Neutron Deficient						
¹⁰⁰ Pd	28			0.019 ± 0.05	109 ± 25	
¹⁰¹ Pd	37			0.025 ± 0.04	145 ± 22	
¹⁰³ Pd	32			0.022 ± 0.01	129 ± 3	200 ± 10
¹⁰³ Ag	29			0.021 ± 0.03	119 ± 18	
¹⁰⁵ Ag	37			0.024 ± 0.06	137 ± 34	
¹¹¹ In	25			0.022 ± 0.00	126 ± 7	

^a 450 MeV proton bombardment

Table VI

Average Projected Ranges Calculated for Spallation

Z	A	Initial conditions			Projected ranges	
		E* (MeV)	$v_{//}$ (MeV/amu) ^{1/2}	$v_{\perp}$ (MeV/amu) ^{1/2}	FW mg/cm ²	BW mg/cm ²
82	203	1800	0.26	0.65	2.0	0.07
82	203	1800	0.17	0.48	1.3	0.2
Experimental values					1.3±0.1	1.1±0.1

Figures

Fig. 1 - Charge dispersion for products with $100 \leq A \leq 115$ from the interaction of ^{238}U with 11.5 GeV protons. Solid curve - overall dispersion curve calculated for $A = 111$; dot-dashed extensions - resolution of solid curve into two components; dashed curve - cumulative cross sections based on the charge dispersion curve. $\odot$ - experimental cumulative yields; $\circ$ - experimental independent yields.

Fig. 2 - Comparison of the charge dispersion for products with $100 \leq A \leq 115$ from the interaction of ^{238}U with 11.5 (solid curve) and 0.45 GeV (dashed curve) protons.

Fig. 3 - Isotopic distribution of neutron deficient palladium nuclides. Solid curve - 11.5 GeV protons on ^{238}U ; dashed curve - 400 MeV protons on silver; dot-dashed curve - fission of nuclide with $Z \sim 70$, $A \sim 170$ by 450 MeV protons as extrapolated from results for ^{238}U and ^{208}Pb . Experimental points are shown whenever available.

Fig. 4 - Dependence of $2W(F+B)$ on effective N/Z of product. The solid curve is constructed from the indicated ranges for the two types of products weighted by their respective cross sections at each N/Z . $\odot$ - Experimental values at 11.5 GeV; $\ominus$ - experimental values at 400 MeV.