Recoil Studies of High-Energy Proton Reactions in Bismuth*

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The ranges in bismuth of Sr^{91}, Ba^{129}, and Ba^{133m} recoil fragments from bombardment of bismuth with protons of 50 Mev to 2.2 Bev have been measured. The kinetic energies of the fragments have been calculated from the measured ranges and other reported range-energy data for fission fragments. The kinetic energy of the strontium recoils for all energies, and that of the barium recoils up to 450 Mev, can be explained as resulting from high-energy fission where most of the mass of the target is divided between two heavy fragments. At 2.2 Bev, the kinetic energy of the bariums recoils is substantially smaller, consistent with a complimentary fragment of about 20 mass units.

The energy deposition in the target nucleus for processes leading to strontium and barium nuclides was obtained from the ratio of recoils projected forward to those projected backward, and subsidiary calculations involving momentum and energy conservation. The calculated values of the energy deposited are substantially lower than the bombarding proton energies, and even somewhat lower than energy values calculated for production of the "fissioning nucleus" from radiochemical data.

Recoil experiments on spallation products, bismuth, lead, and thallium, of masses 198 to 203, from high-energy proton bombardment of bismuth, indicate that thallium nuclides formed *directly* are produced in processes involving alpha-particle emission.

I. INTRODUCTION

THIS investigation, the radiochemical study of recoil phenomena in the high-energy proton bombardment of bismuth, was initiated in order to interpret some observations on the yields of products from the bombardment at the Brookhaven Cosmotron of bismuth and lead with 2.2-Bev protons.[1] The yield values of selected nuclides were measured and, when plotted as a function of mass, showed no indication of a "hump" in the curve corresponding to the fission process. The cross-section curve appeared to decrease monotonically through the mass region usually ascribed to "fission" in the hundred Mev range. The cross sections of neutron deficient nuclides of masses in the range 149 to 128 were found to be about 100-fold larger at 2 Bev than at ~400 Mev,[2,3] whereas those of neutron excessive nuclides of masses 90 to 80 were about 3- to 10-fold smaller.

Because of the shape of the yield-mass curve at 2.2 Bev, it was suspected that the mechanism of the reaction leading to the products in high yield of mass number about 130 might be different from that leading to products of mass about 90. Experiments were designed to test this postulate by studying the ranges of the recoil fragments, the difference between the forward and backward ranges as a measure of the velocity imparted to the target nucleus, and the quantities derived from these measurements, such as, the kinetic energies of the fragments, the kinetic energy of the target nucleus, the energy imparted to the target nucleus as excitation energy, etc. Previous use of the recoil technique for the study of the mechanism of high-energy reactions was made by Fung and Perlman[4] in the case of the production of Na^{24} from Al^{27}, and by Fung and Turkevich[5] in the case of the production of Ni^{65} from Cu^{65} by the $(p,p\pi^+)$ reaction. Earlier recoil work[6–8] on fission fragments from the slow-neutron fission of U^{235} and Pu^{239} showed a decided range-mass number dependence useful for mass-number determination,[6,9] consistent[6] with the theoretically derived range-energy dependence.[10]

The information available on the kinetic energies of the fragments, or their ranges, from fission of heavy elements with high-energy projectiles is indeed sparse. An ionization chamber experiment was performed[11] on bismuth, thorium, and uranium with 45-Mev and 90-Mev neutrons, and it was shown that the mean kinetic energy of the U^{235} fragments was only slightly higher than in the case of slow neutron fission. The excitation energy deposited by the projectile is dissi-

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[3] P. Kruger and N. Sugarman, Phys. Rev. **99**, 1459 (1955).

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[10] N. Bohr, Phys. Rev. **59**, 270 (1941).

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pated by evaporation of particles, mostly neutrons, from the target nucleus, as suggested from the results of early radiochemical studies.[12] Range studies of 18-Mev deuteron and 335-Mev proton fission fragments of uranium were made by Douthett and Templeton[13] who found smaller ranges from 335-Mev proton fission than from 18-Mev deuteron fission. Douthett and Templeton interpreted the differences in ranges of the nuclides studied at the two energies as evidence for the formation of the different fission products with 335-Mev protons from different fissioning nuclei.

The recoil experiments reported here on bismuth were done with protons of energy 50 Mev to 450 Mev at the University of Chicago synchrocyclotron,§ and with 2.2-Bev protons at the Brookhaven Cosmotron.[1] A comparison of the ranges and associated data from recoil investigation in the hundred-Mev region with those found at 2.2 Bev for the product studied should elucidate the formation mechanism at the higher energy. In the hundred-Mev range, extensive radiochemical yield determinations on products from heavy elements have been reported[2,3,14–16] in which the masses in the region 60 to 140 have been assigned to products of fission. Some recoil work was also done with 450-Mev protons on products of mass close to that of the target, spallation products, from which some information on the mechanism of the nuclear reactions leading to these products was obtained.

II. EXPERIMENTAL PROCEDURE

A. Target and Recoil Assembly

The bismuth foil used for the target was made by electroplating bismuth on both sides of a 1-mil electrolytic copper foil. For the synchrocyclotron experiments, the surface coated was about 12 cm² in area, from which an inside piece of 4 cm² was cut out. The bismuth was deposited to a thickness of about 15 to 20 mg/cm² on each side of the copper foil, so that the effective total bismuth thickness was 30 to 40 mg/cm². The weight of bismuth deposited was determined by weighing the 4-cm² piece of the target and subtracting the weight of the copper. The thickness of bismuth varied by about 10% over the 4-cm² area. For the Cosmotron experiments, the targets were considerably larger, $1\frac{1}{4}$ in.$\times 2\frac{1}{2}$ in., with about 23 mg/cm² of bismuth on each side of the copper foil.

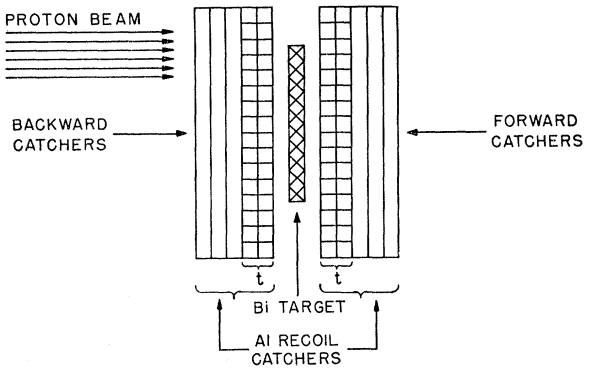

FIG. 1. Target and recoil catcher assembly. t denotes any thickness in the absorber beyond which the recoils are measured [see Eq. (1)].

The recoil catchers used were aluminum foils of thicknesses 0.2 to 4.5 mg/cm². In experiments in which the recoil ranges in aluminum were determined, i.e., "differential" experiments, foils of varying thickness were used and the activity in each was determined. When the total recoil activity leaving the target was determined, i.e., "integral" experiments, only one catcher foil of thickness greater than the range was used. The recoil catchers were mounted adjacent to the target foil, and protruding beyond the target, so that all recoils leaving the target should have been caught in the recoil catchers (Fig. 1). Extra aluminum foils beyond the range of the recoils were included for determination of the activity level from impurity activation.

The target and catcher assembly was mounted in an appropriate target holder for proton bombardment with the internal circulating beam. Energy variation at the synchrocyclotron was effected by variation of the target distance from the center of the machine. Recoils projected along with, opposed to, or perpendicular to the proton beam were collected. After the bombardment, the target and catcher foils were separately prepared for chemical analysis by dissolving in the appropriate acid.

B. Chemical Separations and Radioactivity Measurement

Aliquots of the target and recoil catcher solutions were taken and subjected to radiochemical analysis for the desired elements. In most of the experiments, the solutions were analyzed for strontium and barium; in a few experiments analyses for lead and thallium were also made. The chemical procedures used were essentially the same as those given elsewhere.[17,18] Analyses

[12] R. H. Goeckermann and I. Perlman, Phys. Rev. **76**, 628 (1949).
[13] E. M. Douthett and D. H. Templeton, Phys. Rev. **94**, 128 (1954).
§ *Note added in proof.*—Since this paper was sent to the Editor, a report of Russian work at the Moscow Conference of Academy of Sciences U.S.S.R. (July 1–5, 1955) on high-energy proton fission of uranium, bismuth, and tungsten has come to our attention. Perfilov, Ivanova, Lozhkin, Ostroumov, and Shamov, using photographic plate techniques, present results on the ranges of fission fragments, the asymmetry in mass of the fission process, the energy of excitation for the fission process, etc. The results of this work compare favorably with those given in this paper.
[14] L. Jodra and N. Sugarman, Phys. Rev. **99**, 1470 (1955).
[15] W. E. Nervick and G. T. Seaborg, Phys. Rev. **97**, 1092 (1955).
[16] Folger, Stevenson, and Seaborg, Phys. Rev. **98**, 107 (1955).
[17] Selected papers of Part VI, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Co., Inc., New York, 1951), National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.
[18] W. W. Meinke, University of California Radiation Laboratory Report, UCRL 432, 1949 (unpublished).

of duplicate samples of the same solution, when made, gave agreement to within 2%.

The samples were prepared in the form of precipitates, filtered onto paper disks, mounted on $\frac{1}{16}$-in. aluminum cards, and covered with cellophane. The activity measurements were made with end-window methane-flow proportional counters[19] operating at about 4000 v, or P-10 (90% argon—10% methane) flow counters operating at about 2000 v, with an over-all counting efficiency for beta radiation of about 45%. Throughout the course of these experiments, which lasted for about 1.5 yr, minor electronic adjustments maintained the "standard" counting rate of a U_3O_8 standard to within 0.5%.

The identification of the radioactive species present in the isolated samples was made by analysis of the decay curves and comparison of the half-lives of the analyzed components with reported values.[20] In all cases, the decay curves were consistent with known species.

III. RESULTS. "FISSION" PRODUCTS

A. General Results

The two elements separated in the "fission" region were strontium and barium. The nuclide present in highest intensity in the strontium decay curves at 9 hr after the end of a 0.5-hr bombardment was 9.7-hr Sr^{91}, with some contribution from the 2.7-hr Sr^{92}—3.5-hr Y^{92} decay chain. The contribution of 53-day Sr^{89} was small. At all proton energies, although the shape of the strontium decay curves changed somewhat with energy, the decay curves from the recoil catcher samples were parallel to those from the bismuth target samples to within 1% for 2 days following the bombardment. The correction for strontium activity from impurity activation in the catcher foils was always less than 1% of the recoil activity.

The barium decay curves consisted mostly of 2-hr Ba^{129} and 39-hr Ba^{133m}, with about equal intensities of each at 7 hr after the end of a 0.5-hr bombardment with 450-Mev protons. For lower energy protons, the contribution of Ba^{129} decreased, such that at 50 Mev it was barely discernible. At 2.2 Bev, the Ba^{129} contribution was decidedly enhanced. Under all circumstances, the decay curves of the barium samples from the recoil fractions were parallel to those from the bismuth target to within 3% for the first few days following the bombardment. The contribution from activation of impurities in the aluminum to the recoil barium activity in the catcher foils was as high as 3% in some cases, and was subtracted from the recoil activity.

[19] N. Sugarman and A. Haber, Phys. Rev. **92**, 730 (1953).

[20] Way, Fano, Scott, and Thew, National Bureau of Standards Circular No. 499 (U. S. Government Printing Office, Washington, D. C., 1950) and Supplements; Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

B. Differential Recoil Experiments. Range Determination in Aluminum

The ranges in aluminum of the strontium and barium fragments were determined by analysis of the recoil activity in aluminum recoil catchers. Three recoil catchers of total thickness greater than the range of the fragments were used as catchers for forward and backward recoils in each experiment. Five experiments were performed with 450-Mev protons and two with 2.2-Bev protons. The range of a fragment in a recoil catcher assembly from a thick target is obtained from an analysis in which the following assumptions are made. (1) The fission process occurs by the collision of the proton with the target nucleus, imparting to it a velocity v in the laboratory system along the direction of the proton. (A perpendicular component of velocity does not affect this analysis.) (2) The breakup of the nucleus into fragments is isotropic in the system of the moving target nucleus, with a velocity V for the large fragments, such as strontium or barium. And, (3) the range of a fragment is proportional to its initial velocity. The absorption behavior for recoils is then given by[21]

$$N(t)=\frac{R_0 D}{4}\left[1\pm\eta-\frac{t}{R_0'}\right]^2, \tag{1}$$

where, $N(t)$ is the number of recoils penetrating beyond thickness t in the recoil absorber, D is the number of reactions occurring in the target per mg/cm², R_0 is the range the recoils would have in the *target* material from a stationary target nucleus, R_0' is the corresponding range of the recoils in the *absorber* material, and η is the ratio of the velocity of the target nucleus to that of the fragment in the system of the moving target nucleus (v/V). The plus and minus signs before η are for the cases of forward and backward recoils, respectively.

The absorption expression for $N(t)$ may be put into a reduced form by normalizing $N(t)$ to unity at t equal to zero. The reduced absorption relation is

$$F(t)=\left[1-\frac{t}{R_0'(1\pm\eta)}\right]^2. \tag{2}$$

The absorption behavior for fragments from a moving target nucleus is, then, the same as that for a stationary

TABLE I. Range determinations in aluminum from absorption measurements.

Nuclide	"Forward range," mg/cm² Al		"Backward range," mg/cm² Al		R_0', mg/cm² Al	
	450 Mev	2.2 Bev	450 Mev	2.2 Bev	450 Mev	2.2 Bev
Sr^{91}	3.5±0.1	3.3	3.1±0.1	3.1	3.3±0.1	~3.2
$Ba^{129,133m}$	2.4±0.1	~1.5	1.8±0.05	<1.5	2.1±0.1	<1.5

[21] The derivation of this equation is essentially the same as that for the case of $\eta=0$ except that the thickness t in the absorber is effectively smaller for forward recoils of the amount $\eta R_0'$, and correspondingly larger for backward recoils by the same amount.

one, except that the range in the absorber is replaced by two quantities, a "forward range" equal to $R_0'(1+\eta)$, and a "backward range" equal to $R_0'(1-\eta)$. The range, R_0', is the average of the "forward and backward ranges." The results of the analysis of the differential experiments with 450-Mev protons and 2.2-Bev protons are given in Table I.

The results for 450-Mev protons are averages of at least three of the five total experiments, and those for 2.2-Bev protons represent results of two experiments, one of which was only exploratory. Isolation of Br^{80m} from the first 2.2-Bev proton experiment gave results almost identical to those of Sr^{91}. We note that the range, R_0', of strontium recoils is about the same for 450-Mev and 2.2-Bev proton bombardment, whereas that of barium recoils, is substantially smaller at 2.2 Bev.

A plot of the reduced absorption curve for strontium recoils from 450-Mev proton bombardment, plotting both forward and backward absorption data on the same graph, with the ranges 3.5 mg/cm² Al for the "forward range" and 3.1 mg/cm² Al for the "backward range," is given in Fig. 2. The smooth curve through the points is the quadratic relation given in Eq. (2). For comparison, there is also included in Fig. 2 a plot of absorption data for Sr^{89} recoils from slow neutron fission of a thin target of uranium from other work,[8] where the expected linear absorption behavior is observed. The agreement of the points for the thick target to a quadratic curve for values of $t/R_0'(1\pm\eta)$ to 0.7 is indeed gratifying, and indicates that within the limits of this analysis the assumptions stated earlier are satisfied.[22]

The absorption data may be used to solve for η by the equation

$$\eta=\frac{\text{"forward range"}-\text{"backward range"}}{\text{"forward range"}+\text{"backward range"}}. \quad (3)$$

Since small errors in the "forward or backward ranges" will appear as sizeable errors in η, only the 450-Mev data may be subjected to this treatment. The values of η are 0.061 ± 0.02 for strontium and 0.14 ± 0.03 for barium, the large errors in η arising from the relatively small (less than 5%) errors in the range data. A discussion of the importance of η in the study of the mechanism of the nuclear reaction will be deferred until after better values are presented from the integral experiments (Sec. III, C).

[22] Recent work by R. L. Wolke [Phys. Rev. **98**, 1199 (1955) and University of Chicago (unpublished)] shows that the Sr^{91} recoils formed from bismuth with 450 Mev protons are distributed *anisotropically* in the system of the struck target nucleus. The distribution in angle is given by $a+b\cos^2\theta$, where θ is the angle between the direction of the proton beam and the fragment in the system of the struck target nucleus. The ratio b/a is approximately 0.15. The values of η, R_0, v, etc. given later are not changed appreciably by corrections for anisotropy.

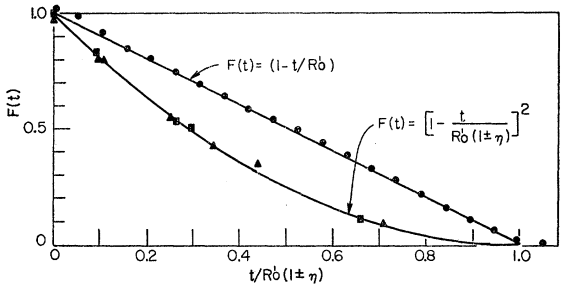

FIG. 2. Reduced recoil absorption curves for thin and thick targets. ● Sr^{89} from slow neutron fission of U^{235} (*thin* target; $R_0'=3.74$ mg/cm² Al); ■, Sr^{91} forward recoils, and ▲, Sr^{91} backward recoils, from 450-Mev proton bombardment of Bi^{209} (*thick* target, "forward range"=3.5 mg/cm² Al; "backward range"=3.1 mg/cm² Al).

C. Integral Recoil Experiments. Range Determinations in Bismuth

Since it was shown in the differential recoil experiments that the recoiling fragments satisfied the predicted quadratic behavior, it was clear that range data (in bismuth), and associated quantities, such as η, v, V, etc. could be determined more accurately from integral experiments than from differential ones because of the inherently higher precision possible. In these experiments, the target foil was surrounded by a heavy recoil catcher, ~4.5 mg/cm², sufficiently thick to stop all the recoils. The total recoils projected forward, backward, or perpendicular to the direction of the protons were thus absorbed. By analysis of the forward and backward recoil activity and of the residual activity in the target, calculations of η, R_0, etc. could be made. From Eq. (1) it is seen that the ratio of total forward recoils (ReF) to total backward recoils (ReB) is given by

$$\frac{\text{ReF}}{\text{ReB}}=\left[\frac{1+\eta}{1-\eta}\right]^2, \quad \text{and} \quad \eta=\frac{(\text{ReF}/\text{ReB})^{\frac{1}{2}}-1}{(\text{ReF}/\text{ReB})^{\frac{1}{2}}+1}. \quad (4)$$

Also, since ReF is related to the range in bismuth, R_0, by the following expression:

$$\text{ReF}=\tfrac{1}{4}R_0D(1+\eta)^2, \quad (5)$$

and ReB is similarly related except for the sign before η, then the fraction of the total activity found in the forward or backward recoils from a target of thickness W, in mg/cm², is given by

$$\text{fraction}\begin{Bmatrix}\text{forward}\\ \text{or}\\ \text{backward}\end{Bmatrix}=\frac{\text{ReF or ReB}}{WD}=\frac{R_0}{4W}(1\pm\eta)^2, \quad (6)$$

from which R_0 can be shown to be

$$R_0=\left(4W\times\text{fraction}\begin{Bmatrix}\text{forward}\\ \text{or}\\ \text{backward}\end{Bmatrix}\right)\Big/(1\pm\eta)^2. \quad (7)$$

The determination to high precision of ReF, ReB, and their ratio is, then, necessary for accurate deter-

TABLE II. Results of typical recoil experiment (450-Mev protons; target thickness, 32 mg/cm² Bi).

Nuclide isolated	Sample	Activity corr. for aliquot and yield counts/min	Total activity counts/min	Fraction of total recoiling out	ReF/ReB	η	R_0, mg/cm² Bi
Sr^{91}	Target	83 000					
	Forward recoils, ReF	7850	97 410	0.0806	1.20	0.0450	9.4
	Backward recoils, ReB	6560		0.0674			
$Ba^{129,133m}$	Target	2860					
	Forward recoils, ReF	189	3170	0.0596	1.56	0.111	6.2
	Backward recoils, ReB	121		0.0382			

mination of η, R_0, and the derived quantities such as kinetic energies of the fragments, etc. The results of a typical integral recoil experiment at 450 Mev, showing the nature of the results and calculations, are given in Table II. A summary of the results on ReF/ReB of strontium and barium recoils for all the experiments performed with protons of 50-Mev to 2.2-Bev energy is given in Table III. The results are the averages of three to five experiments at each energy, except for the 450-Mev proton data which are averages of seven experiments, and the 2.2-Bev proton data which represent only one experiment and for which no errors are quoted. The errors, where given, are the average deviations from the mean. The value of η for each energy is calculated from the average ReF/ReB at that energy as given by Eq. (4). The range value, R_0, at each energy is calculated by use of Eq. (7) from the average value of the product of W and the fraction of activity forward or backward for each experiment. The last two columns of Table III give a quantity proportional to v from the product of η and R_0. The significance of this quantity in the interpretation of the mechanism of the nuclear reaction will be discussed later. No data at 50 Mev on barium are given in Table III since the activity of barium was hardly discernible at this energy. The cross section for formation of barium decreases rapidly as the proton energy is decreased because of the low neutron to proton ratio[14] of the barium nuclides.

Despite the relatively small error in ReF/ReB of about 2%, the error in η is about 10%. Still, the η values determined from these integral recoil experiments are considerably more precise than those obtained from the differential recoil experiments. The η values for 450 Mev protons from the integral recoil experiments agree within the limits of error with the η results given earlier in Sec. III, B. The error in R_0 arises partly from the error in determining the fraction of recoils and partly from the nonuniformity of the target foil. Since the proton beam is not uniform over the target foil, varying by a factor of two in intensity every 2 or 3 mm in the radial dimension of the machine, the section of highest intensity may be relatively thinner or thicker than the average thickness, W, and hence give a relatively larger or smaller range, respectively. It may be noted that the relative errors in R_0 are larger than those in ReF/ReB for this reason. Derived quantities, such as v, etc., have even larger relative errors.

A cursory examination of the results of Table III shows that no serious change in any of the quantities occurs in the energy range 100 Mev to 2.2 Bev for Sr^{91} recoils. This result would indicate that the nature of the process producing this nuclide, namely fission, is probably the same throughout this energy range. In the case of $Ba^{129,133m}$, however, large changes are noted in the various quantities of Table III in going from 450 Mev to 2.2 Bev, as though a change in the nature of the process producing this nuclide were occurring.

The range data for strontium and barium recoils given in Table III may be used to calculate the kinetic energies of the fragments for the various proton energies by use of the range data in aluminum of Douthett and Templeton.[13] On the assumption that for a given mass the range is proportional to the initial velocity of the fragment, the proportionality constant in the range-velocity or range-energy relations may be calculated. The ranges of Sr^{91} and Ba^{140} recoils from 18-Mev deuteron bombardment of uranium are 4.20 mg/cm² Al and 3.10 mg/cm² Al, respectively. We can solve for the

TABLE III. Summary of recoil results.

Proton energy Mev	ReF/ReB		η		R_0, mg/cm² Bi		$\eta \times R_0$	
	Sr^{91}	$Ba^{129,133m}$	Sr^{91}	$Ba^{129,133m}$	Sr^{91}	$Ba^{129,133m}$	Sr^{91}	$Ba^{129,133m}$
50	1.10±0.02	···	0.023±0.005	···	9.1±0.4	···	0.21±0.04	···
100	1.17±0.02	1.31±0.04	0.039±0.005	0.066±0.01	10.8±0.4	6.8±0.3	0.42±0.06	0.45±0.07
180	1.20±0.02	1.45±0.02	0.045±0.005	0.092±0.005	10.1±0.2	6.4±0.2	0.45±0.05	0.59±0.04
300	1.20±0.03	1.42±0.02	0.045±0.007	0.087±0.005	9.6±0.3	6.1±0.1	0.43±0.07	0.53±0.03
450	1.20±0.02	1.55±0.02	0.045±0.005	0.109±0.005	9.5±0.2	5.9±0.2	0.43±0.05	0.64±0.04
450	[a]	[a]	[a]	[a]	9.5±0.2[a]	5.8±0.1[a]		
2200	1.24	3.8	0.053	0.32	10.0	3.0	0.53	0.96

[a] Perpendicular recoils collected; only R_0 measured. Average of two experiments.

proportionality constant, k, in the range-energy expression

$$E=kR_0^2, \quad (8)$$

where E is the initial kinetic energy of the fragment in Mev and R_0 is the range in bismuth in mg/cm^2, using 168 Mev as the average total kinetic energy of fission, and 2.9 as the ratio for ranges in bismuth compared to aluminum.[23] This ratio is obtained by comparing the 450-Mev range values of Table III with the range values given in Table I. The values of k so obtained are 0.70 for Sr^{91} and 0.80 for Ba^{140}.

The kinetic energies of the Sr^{91} and $Ba^{129,133m}$ fragments may be calculated from the range data using the same value of k for $Ba^{129,133m}$ as for Ba^{140}. The results are given in Table IV. The kinetic energy of the strontium recoils rises from 58 Mev to 82 Mev as the proton energy increases from 50 Mev to 100 Mev, then decreases slowly from 82 Mev to 63 Mev for increasing proton energy to 450 Mev. For protons of 2.2 Bev, approximately the same fragment kinetic energy is observed

TABLE IV. Energy calculations for Sr^{91} and $Ba^{129,133m}$ recoils from range data.

Proton energy, Mev	R_0(Sr)	E(Sr),[a] Mev	R_0(Ba)	E(Ba),[b] Mev	R_0(Sr)[c] / R_0(Ba)	E(Sr)[d] / E(Ba)
50	9.1±0.4	58±5	...	...	...	...
100	10.8±0.4	82±6	6.8±0.3	37±3	1.60±0.03	2.24±0.09
180	10.1±0.2	71±3	6.4±0.2	33±2	1.58±0.01	2.19±0.04
300	9.6±0.3	65±4	6.1±0.1	30±1	1.58±0.02	2.19±0.07
450	9.5±0.2	63±2	5.9±0.2	28±2	1.61±0.05	2.28±0.13
2200	10.0	70	3.0	7	3.3	9.5

[a] $E(\mathrm{Sr})=0.70\,R_0(\mathrm{Sr})^2$.
[b] $E(\mathrm{Ba})=0.80\,R_0(\mathrm{Ba})^2$.
[c] $R_0(\mathrm{Sr})/R_0(\mathrm{Ba})$ is obtained from the average of the ratio of the fraction of strontium activity recoiling out of the target to the fraction of barium recoils for each experiment, thus avoiding the use of the somewhat uncertain target thickness, W.
[d] $E(\mathrm{Sr})/E(\mathrm{Ba})$ is obtained from $R_0(\mathrm{Sr})/R_0(\mathrm{Ba})$ by the equation: $E(\mathrm{Sr})/E(\mathrm{Ba})=(0.70/0.80)[R_0(\mathrm{Sr})/R_0(\mathrm{Ba})]^2$.

(Fig. 3). The kinetic energy of the barium recoils shows the same trend as that of Sr^{91} in the 100 Mev to 450-Mev proton energy range, decreasing from a value of 37 Mev to 28 Mev, such that the ratio of the kinetic energy of the strontium recoils to that of the barium recoils, $E(\mathrm{Sr})/E(\mathrm{Ba})$, is roughly constant at about 2.22. For 2.2-Bev protons, however, the kinetic energy of the barium recoils is substantially lower, 7 Mev, and the ratio $E(\mathrm{Sr})/E(\mathrm{Ba})$ is 9.5. Any deviation from the linear range *vs* velocity dependence at these low energies will make the energy values higher than those calculated. The only experimental value for comparison of the kinetic energies of Table IV with published work is the comparison of the (82±6) Mev value for Sr^{91} recoils from 100 Mev proton bombardment, with the most probable kinetic energy value of 75 Mev found from 90 Mev neutron bombardment of bismuth, as measured in ionization chamber experiments.[11] Since Sr^{91} would be expected to be lighter than the most probable fission

[23] E. Segrè and C. Wiegand [Phys. Rev. **70**, 808 (1946)] give a value of 3 for the ratio of the mass stopping power of aluminum to that of gold for gross fission recoils.

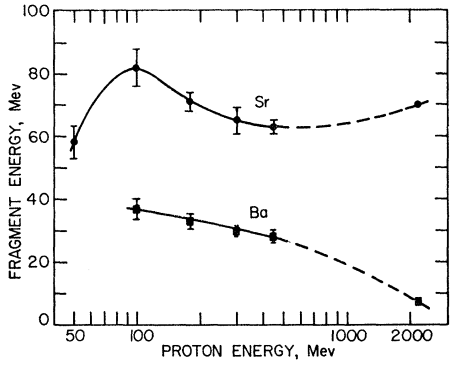

FIG. 3. Kinetic energies of Sr^{91} and $Ba^{129,133m}$ recoil fragments as determined from range measurements.

fragment for 100-Mev proton fission,[14] the most probable kinetic energy should be some 10% lower than 82 Mev, making the agreement with the ionization chamber value even better.

IV. DISCUSSION OF "FISSION" RESULTS

A. Comparison with Kinetic Energies Calculated from Coulombic Repulsion

The kinetic energies of the recoil fragments may be calculated on the basis of the following model. The bombarding proton despoits some of its energy in passing through the nucleus. If the excitation energy is dissipated by evaporation of particles before fission, then the resultant de-excited nucleus of lower neutron to proton ratio is the "fissioning" nucleus to be used in the calculation. On the other hand, if the fission process precedes particle evaporation, then the "fissioning" nucleus to be used for the calculation is the target nucleus, or one near it in mass and charge, because of "knock-on" particles.[24] Particle evaporation occurs from the fragments thereby lowering their masses and kinetic energies. The first assumption on the "fissioning" nucleus was used in the calculation, namely evaporation of particles prior to fission. The kinetic energies of the strontium and barium fragments were calculated on the basis of two contiguous spherical balls, of radius $1.45\times10^{-13}A^{\frac{1}{3}}$ cm, using for the "fissioning" nuclei at the various energies the results of previous radiochemical observations.[3,14] The calculated values for Sr^{91} decrease slowly from 104 Mev for 50-Mev protons, using $_{84}Po^{205}$ as the fissioning nucleus,[25] to 91 Mev for 450-Mev protons, using $_{80}Hg^{186}$ as the fissioning nucleus. For $Ba^{129,133m}$, the calculated values decrease from 58 Mev for 100-Mev protons, using $_{83}Bi^{200}$ as the fissioning nucleus, to 46 Mev for 450-Mev protons, using $_{80}Hg^{186}$

[24] R. Serber, Phys. Rev. **72**, 1114 (1947); M. L. Goldberger, Phys. Rev. **74**, 1269 (1948); Bernardini, Booth, and Lindenbaum, Phys. Rev. **85**, 826 (1952).

[25] At the 50-Mev and 100-Mev proton energies, the fissioning nuclei used in the calculations are somewhat higher in mass and charge than those given in reference 14. The calculated results are not sensitive to small changes in mass and charge of the fissioning nucleus.

TABLE V. Results of calculations on kinetic energy and energy deposition in struck nucleus. (Separate calculations made from strontium and barium data.)

Proton energy, Mev	Velocity of struck nucleus, v [a]		Kinetic energy of struck nucleus, Mev		Energy deposited, Mev	
	Sr^{91}	$Ba^{129,133m}$	Sr^{91}	$Ba^{129,133m}$	Sr^{91}	$Ba^{129,133m}$
50	0.026±0.005		0.071±0.028		40±10	
100	0.052±0.008	0.052±0.008	0.28±0.09	0.28±0.09	94±6	95±6
180	0.056±0.006	0.068±0.004	0.33±0.07	0.49±0.06	147±10	164±10
300	0.053±0.009	0.061±0.003	0.30±0.10	0.39±0.04	190±20	212±10
450	0.053±0.006	0.073±0.004	0.30±0.07	0.56±0.06	226±20	290±15
2200	0.066	0.11	0.46	1.27	400	660

[a] Units of v are (Mev/nucleon mass)$^{\frac{1}{2}}$.

as the fissioning nucleus. Calculation of the kinetic energies on the assumption of evaporation of particles following fission, with the excitation energy proportional to the mass of the fragment, leads to values about 5% lower.

Three general observations on the comparison of the calculated kinetic energies with the observed ones should be made. First, the calculated values are considerably higher than those observed, the ratio between them varying from 1.3 to 1.8. This result is not surprising in view of the fact that even for slow neutron fission of U^{235} the values calculated on the basis of the spherical ball model are about 20% higher than the observed fragment energies. The fact that the ratio of the calculated to the observed kinetic energy is on the average about 40% higher for proton fission of bismuth than for slow neutron fission of U^{235} must mean that the distortion from the spherical ball model for high energy fission of bismuth is even more pronounced than in the low-energy uranium case.[26] Secondly, the calculated kinetic energy for Sr^{91} is 104 Mev for 50-Mev protons and 103 Mev for 100-Mev protons, whereas the observed values are (58±5) Mev and (82±6) Mev, respectively. Apparently, there is some marked change in the fission process occurring in this energy interval not accounted for in the calculation. It is unlikely that the energy uncertainty in the proton beam at 50 Mev could produce this effect, since the presence of low energy protons in large intensity, if they exist, would be expected to give larger rather than smaller ranges, or kinetic energies. Thirdly, the ratio of the kinetic energies of the strontium fragments to those of barium fragments is constant at a value of 2.22±0.04 in the 100 Mev to 450-Mev energy interval. The calculated ratio, on the other hand, increases almost linearly from 1.78 at 100 Mev to 1.98 at 450 Mev (Fig. 4). The difference in behavior between the observed and calculated ratios of kinetic energy, although apparently outside the experimental error, is perhaps not too secure.

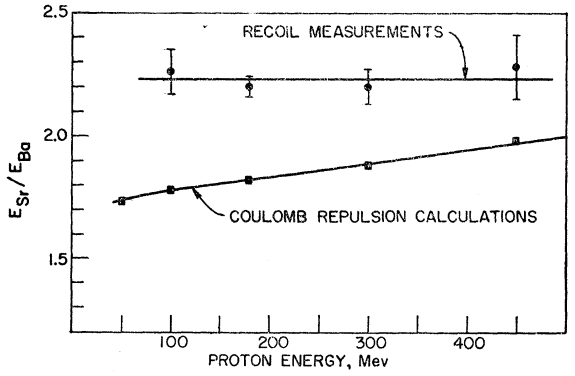

FIG. 4. Comparison of ratios of kinetic energies of strontium fragments to those of barium, $E(\mathrm{Sr})/E(\mathrm{Ba})$, from recoil measurements with those calculated from Coulombic repulsion.

[26] S. Frankel and N. Metropolis, Phys. Rev. **72**, 914 (1947).

B. Calculation of Energy Transferred to Target Nucleus

The energy transferred to the target nucleus may be calculated from the measured values of η and the constants relating range and energy used earlier. Although the linear relationship of range to velocity implied in these calculations is not expected to obtain at values of the velocity of the order of that of the struck target nucleus, the treatment used here avoids this difficulty since the calculations are made for values of the velocity in the range of the fragment velocity. Table III gives values proportional to v in the last two columns from the products of η and R_0. From the proportionality constants relating range and energy derived earlier, it can be shown that $R_0(\mathrm{Sr})=8.07V_{\mathrm{Sr}}$, and $R_0(\mathrm{Ba})=8.70V_{\mathrm{Ba}}$, where R_0 is given in mg/cm^2 Bi, and V_{Sr} or V_{Ba} are the fragment velocities in the system of the moving target nucleus. The units of V are (Mev/nucleon mass)$^{\frac{1}{2}}$. For Sr^{91} recoils, then, $\eta\times R_0=8.07v$ and for $Ba^{129,133m}$ recoils, $\eta\times R_0=8.70v$. The units of v are the same as those of V. The results on v calculated separately from the strontium and barium data are given in Table V in columns 2 and 3. It should be mentioned that the value of v calculated at any energy is the component of the true velocity vector along the direction of the proton beam; the transverse component is not observed in these experiments. The kinetic energy of the struck nucleus is given by $\frac{1}{2}Mv^2$, where M is its mass. Neglecting any "knock-on" particles from the cascade process in the nucleus,[24] which at most will affect the results of the kinetic energy calculation by a few percent, one can calculate the

kinetic energies from the v values, equating the energy to $105v^2$. The values of the kinetic energy are given in columns 4 and 5 of Table V. Since v represents only one component of the velocity, the kinetic energies as calculated are smaller than the true kinetic energies by the square of the cosine of the angle between the direction of the proton beam and the direction of motion of the struck nucleus.

A calculation of the kinetic energy left behind in the struck nucleus by the bombarding proton can be made from conservation of momentum of the system. The two assumptions made are: (1) that v is aligned along the direction of the bombarding proton, and (2) that only one high energy particle leaves during the cascade process, the other "knock-on" particles being of much lower energy. The calculated values of the energy deposition for the production of Sr^{91} and $Ba^{129,133m}$ for the various proton energies are given in columns 6 and 7 of Table V.

A comparison of the calculated results given in Table V from the strontium data with those from the barium data shows that there is good agreement between them for the various quantities for 100-Mev, 180-Mev, and 300-Mev protons. The values of the velocity, v, the kinetic energy of the struck nucleus, and the deposition energy agree to within the experimental errors. This agreement would imply that the production process for the two nuclides can be considered to be the same, namely fission, and that both nuclides are coming from the same fissioning nucleus. For 450-Mev protons, and even more markedly, for 2.2-Bev protons, the strontium values are lower than those of barium, implying a larger transfer of energy to the target nucleus for the production of barium than for strontium. At 450 Mev, the small excess energy transfer needed for production of barium produced no marked change in the trend of the kinetic energy values of the barium recoil fragments, as seen in Table IV and Fig. 3. At 2.2 Bev, there is little question from the results of both Table IV and Table V that the production of $Ba^{129,133m}$ cannot be ascribed to the same fission process producing Sr^{91}. The 7-Mev kinetic energy calculated for the barium recoils at 2.2 Bev, on the assumption of an isotropic distribution in the system of the struck target nucleus, still demands the simultaneous emission of another fragment of mass number about 20 to achieve even this low energy.

The values for the energy deposition given in Table V are uniformly lower than those calculated for production of the "fissioning" nucleus, as determined from radiochemical yield data, on the assumption that all of the particles are emitted by evaporation prior to fission. For example, for 450 Mev protons on bismuth,[3] it was calculated that a minimum energy of 320 Mev is required for formation of the most probable "fissioning" nucleus, $_{80}Hg^{186}$, which is in excess of the average of the values calculated from the strontium and barium data of Table V, namely 260 Mev. Similar calculations on the minimum energies required to produce the "fissioning" nuclei at the lower proton energies[14] again lead to values higher than those of Table V. This discrepancy in the values of the energy deposition may result either from the mode of analysis of the radiochemical data[3] leading to a choice of the "fissioning nucleus" too small in mass and charge, or the assumption that all of the particles lost in producing the "fissioning" nucleus are lost singly and prior to the fission act. Post-fission evaporation of the particles yields a somewhat lower energy value for the energy deposited.

Perhaps noteworthy of general comment is the observation from Table V that the energy deposition for the process under investigation at each energy is smaller than the available energy, a consequence of the transparency of nuclei for high-energy projectiles.[24] Even for 50-Mev protons, where the deposition energy of (40 ± 10) Mev appears to be the same as that available within the limits of error, the collision does not involve transfer of all of the proton energy, as seen from the value of the kinetic energy of the struck nucleus (Table V) which is some threefold lower than that expected for a totally inelastic collision of the proton with the target nucleus.

V. RESULTS AND DISCUSSION OF SPALLATION REACTIONS

Recoil experiments were performed on spallation products from the bombardment of bismuth with 450-Mev protons. Bismuth, lead, and thallium spallation products in the mass number range 198 to 203 were investigated. In the differential recoil experiments it was soon seen that the spallation recoil fragments did not penetrate beyond the thinnest aluminum catcher used, about 0.2 mg/cm^2. The activity found in catchers beyond this thickness could be accounted for by impurity activation. Hence, only information on the total recoil activity found in the forward and backward directions to the proton beam could be obtained.

The bismuth target foil and aluminum recoil catchers were dissolved, and aliquots were taken for lead and thallium analysis. The separations of thallium and lead were appropriately timed in order to give the maximum information possible on individual species in these complex decay chains.[27,28] Although the fraction of activity observed to recoil out of the target was consideraly smaller than for Sr^{91} and $Ba^{129,133m}$, the intensity of activity in the forward recoil catcher was only slightly smaller than that of Sr^{91} because of the high cross sections of the spallation products. The summary of the results of the recoil data from the lead and thallium analyses are given in Table VI. The recoiling nucleus responsible for the radioactivity in the lead samples of the catcher foils was assigned in each case from the known decay chains and formation cross sections[27,29]

[27] W. E. Bennett, Phys. Rev. **94**, 997 (1954).
[28] Bergström, Hill, and de Pasquali, Phys. Rev. **92**, 918 (1953).
[29] M. Campos, University of Chicago (unpublished).

TABLE VI. Recoil results on spallation products from 450-Mev proton bombardment of bismuth.

Mass number	Recoiling nucleus	"Effective range,"[a] mg/cm² Bi Forward	Backward	Recoil ratio, forward/backward
198	Tl	0.10	0.07	1.4
199	Bi	0.08	0.007	12
200	Bi and Pb	0.07	0.007	10
	Tl	0.14	0.09	1.6
201	Bi and Pb	0.05	~0.007	~7
202	Tl	0.11	0.10	1.1
203	Bi	0.045	~0.006	~8
	Pb	0.045	~0.008	~6

[a] The "effective range" is the product of the fraction of activity found in the recoil catchers and the thickness of target in mg/cm².

for the individual members of the decay chains. For mass 199, the cross section for production of bismuth is much higher than that of lead and, hence, the recoiling nucleus is Bi^{199}. For masses 200 and 201, where the formation cross sections of Bi^{200} and Pb^{200}, and Bi^{201} and Pb^{201}, are similar, and where the half-lives of the bismuth parents are short, the recoiling nucleus is mixed Bi—Pb. For mass 203, where the bismuth parent is of 12-hr half-life, and the first separation of lead was made before much of the Bi^{203} had decayed, results were obtained for Bi^{203} and Pb^{203} recoiling nuclei separately. For thallium, the first separation was done within 1 hr after the end of bombardment, when little thallium had grown in from lead and bismuth, and so the recoiling activity, and that in the target, represent the thallium formed *directly*.

The quantity used for describing the recoil behavior of a given spallation nuclide is the "effective range," forward or backward. The "effective range" is the product of the fraction of activity recoiling out of the target multiplied by the target thickness, and, so, is the effective thickness of bismuth responsible for the recoil activity. For the case of isotropic emission of recoils, and a velocity of the struck nucleus small relative to the fragment velocity, the forward and backward "effective ranges" are almost equal, such as was observed for strontium and barium fragments. The range of the fragments in the target material, in this case, is equal to about four times the "effective range." For the case where the fragment velocity, in the system, of the struck nucleus, is of the same order as that of the struck nucleus, the forward "effective range" is many times the backward "effective range," and the range of the fragments in the target material is only slightly larger than the forward "effective range."

The major results of Table VI are the relatively small "effective ranges" for the spallation products, and the large difference in the ratio of forward to backward recoils for bismuth and lead recoils compared to thallium recoils. The bismuth and lead recoil data, of large forward to backward ratio, may then be interpreted as the result of the motion of the struck nucleus, in which the order of 100 Mev of energy is deposited, and in which the loss of particles is almost isotropic in the system of the moving target nucleus. The fragment velocity in this system would, consequently, be small. The backward recoils could be the result of scattering, or of some anisotropy in the momentum balance from the evaporated or "knock-on" particles. This interpretation of the large forward to backward ratio for the bismuth and lead recoil fragments is essentially the same as that used for the explanation of the similar recoil behavior of heavy fragments from other target nuclei bombarded with 340-Mev to 450-Mev protons, e.g., Na^{24} from aluminum,[4] Ni^{57} and Ni^{65} from copper,[5] and Cr^{49} from copper.[30] Although these target nuclei are considerably lighter than bismuth, the essential features of the reactions are the same, namely, the order of 100-Mev energy deposition is required for the reaction, and the velocity of the fragments in the system of the struck nucleus is of the same order as that of the struck nucleus.

It is noted from Table VI that the forward "effective range" of bismuth and lead recoils increases as the number of particles lost increases. This is the expected trend for increasing energy deposition. A value of 0.08 mg/cm² Bi is observed for Bi^{199} compared to 0.045 mg/cm² Bi for Pb^{203} or Bi^{203}. Since the backward "effective range" of these nuclides is roughly constant over this mass range, at a value of about 0.007 mg/cm² Bi, the change in the ratio of the forward to backward recoils from 12 for Bi^{199} to ~6 for Pb^{203} is due mostly to the change in the forward "effective range."

The spallation reactions leading to the bismuth and lead recoils studied should involve energy transfers of the order of 100 Mev to 150 Mev, at least for the lower-mass number species. In these cases, calculations may be made on the expected minimum kinetic energy of the struck nucleus, again on the assumption that the velocity of the struck nucleus is along the direction of the bombarding proton. The kinetic energies of the struck nucleus are in the range 50 kev to 100 kev for energy deposition of 100 Mev to 150 Mev. Kinetic energies of this amount for heavy nuclei are encountered in recoils from alpha-particle emission. The range reported by Hevesy and co-workers[31] for Pb^{212} (ThB) recoils, of about 130-kev kinetic energy, in lead is about 0.06 mg/cm², which, when compared to the forward "effective ranges" for $(Bi+Pb)^{199,200}$ of 0.07–0.08 mg/cm² Bi, is in excellent agreement. We conclude, therefore, that the bismuth and lead spallation products studied are produced from inelastic collisions of the proton with the bismuth nucleus in which relatively small amounts of energy are deposited. The excitation energy of the target nucleus is dissipated by the evaporation of particles.

[30] R. E. Batzel and G. T. Seaborg [Phys. Rev. **82**, 607 (1951)] analyzed their recoil data on Cr^{49} using a velocity of the struck Cu^{63} nucleus about one-half that of the Cr^{49} fragments. Conservation of momentum leads to a minimum energy deposition in the Cu^{63} nucleus amounting to about 200 Mev, which seems high for the simple reaction written for the production of Cr^{49}.

[31] Hevesy, Seith, and Keil, Z. Physik **79**, 197 (1932).

The recoiling thallium nuclides formed *directly* have forward to backward ratios almost equal to unity. Inasmuch as a forward velocity component would be expected from the transfer of momentum to the target nucleus, similar in magnitude to that observed for lead and bismuth recoils, the apparent cancellation of this in the production of thallium nuclides must be the result of another process, isotropic in nature, and capable of transferring appreciable momentum. It is proposed that the thallium nuclides studied are formed by the evaporation of neutrons and an alpha particle following the passage of the bombarding proton through the nucleus. The recoil momentum of the thallium nucleus resulting from an alpha particle emitted with an energy of the order of 30 Mev would be about 2.5 times the momentum of the struck nucleus and would lead to a more isotropic distribution of resultant recoils. The range of the thallium recoils can then be taken to be about four times the "effective range," which would be about 0.4 mg/cm^2 Bi, or about 5 to 6 times the range of the lead and bismuth recoils. The range-velocity dependence for heavy fragments at these low kinetic energies, where the range is varying with the second to third power of the velocity,[32] yields an expected value of about 8 for the ratio of the range of the thallium recoils relative to those of the bismuth and lead recoils, from their velocity ratio of about 2.5. The agreement between the expected and observed values for the ratio of the ranges lends support to the model advanced for the production of the thallium nuclides of mass number $\sim$200, in which an alpha particle and neutrons are emitted. In earlier work on the photoactivation of bismuth with 86 Mev bremsstrahlung,[33] the yield of *directly* formed Tl^{201} was higher than that of *directly* formed Pb^{201}. The formation of Pb^{201} by photons involves the emission of one proton and neutrons, and that of Tl^{201} two protons, or an alpha particle, and neutrons. The fact that the yield of Tl^{201} is higher than the yield of Pb^{201} probably means that in this case, too, the Tl^{201} nuclide formation involves alpha-particle emission.

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[32] J. Knipp and E. Teller, Phys. Rev. **59**, 659 (1941).

[33] N. Sugarman and R. Peters, Phys. Rev. **81**, 951 (1951).

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Meson Production in Nucleon-Nucleon Collisions at High Energies*

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Calculations based on a statistical model have yielded results concerning the relative probabilities for the different multiplicities and charge distribution of mesons produced in nucleon-nucleon collisions. In deviating from a pure statistical model the important effects of final state interactions and various selection rules have been included, using results of meson-nucleon scattering experiments. Notably in the results the suppression of some one-meson final states by consideration of the Pauli principle and conservation of angular momentum and parity, along with the enhancement of two-meson states due to resonance effects, have brought about results which are in closer agreement with experiment than predictions of a pure statistical nature. Account was taken of the final state interactions by considering separately the nucleon-nucleon and meson-nucleon interactions, a separation made plausible by consideration of the small amount of kinetic energy taken away by the more massive particles. Meson-meson interactions were neglected. The nucleon-nucleon interaction was taken care of by introducing in the statistical weight a factor which is the square of the wave function for the scattering of two nucleons evaluated at the origin of their interaction. The meson-nucleon final state scattering was treated by the method discussed by Chew, modified for the case of a meson scattering off two stationary and superposed nucleons. Multiplicities up to two mesons were considered. On comparing with experimental results, at 1.7-Bev bombarding energy of neutrons on protons the ratio of the probability of occurrence of the final states $(np+-):(pp-0):(pp-)$ is calculated to be 3.0:1.0:0.9, while experiment gives 3.3:1:0.8. The ratio of the total probability for double meson production to that for single meson production at this energy is 1.2, while a modified result of observations gives 1.4. Results on proton-proton collisions do not yield good agreement with present observations.

I. INTRODUCTION

IN Fermi's[1,2] statistical theory of multiple meson production, the relative probabilities for alternative processes initiated by a nucleon-nucleon collision depend primarily upon the volume in phase space available to each final state when the energy of the colliding nucleons is high. The dependence upon the dynamics involved is argued to be diminishingly small under the assumption that all possible final states are equally excited due to the strong interactions involved. Thus,

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[1] E. Fermi, Progr. Theoret. Phys. (Japan) **5**, 570 (1950).

[2] E. Fermi, Phys. Rev. **92**, 452 (1953); **93**, 1434 (1954).