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Interaction of Negative Pions with Mercury*

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Radiochemical separation of expected fission products from mercury irradiated with 122-Mev π^- mesons demonstrates that fission occurs. The fission products are distributed in yield similar to those from the fission of mercury by high-energy neutrons. The branching ratio for meson fission is about 0.5 percent. Thallium activities found in the meson-irradiated mercury are explained as produced by secondary fast protons.

I. INTRODUCTION

RADIOCHEMICAL studies of the interaction of negative pions with nuclei are in progress with the meson beams from the University of Chicago synchrocyclotron. Yields of the disintegration products of meson interaction on oxygen and nitrogen,[1] bromine,[2] arsenic,[3] zinc,[4] and iodine,[5] and preliminary studies of meson fission of mercury[6] have been reported. It had been shown earlier, in photographic plate experiments, that meson fission of uranium occurs.[7] Recently the probability for fission of uranium has been shown to be 37 percent[8] for negative pions and 15 percent for muons.

Results of the investigation of the interaction of 122-Mev negative pions with mercury are reported here. Isolation of expected radioactive products from the irradiated mercury demonstrates that fission occurs. It was found that the distribution of fission products is similar to that observed in bombardment of other heavy elements with high-energy particles,[9–11] and that the branching ratio of about 0.5 percent for the fission process is roughly that expected according to the models of meson interaction with nuclei.[12] Thallium activity was also isolated and it was demonstrated that secondary fast protons from pion interactions were responsible for its production.

TABLE I. Ratio of activity in wash solutions.

Element	Nuclide studied	Ratio of 1st to 2nd wash
As	91-min As^{78}	5
Br	4.4-hr Br^{80m}	2–7
Sr	9.7-hr Sr^{91}+2.7-hr Sr^{92}	>700
Y	3.5-hr Y^{92}+10-hr Y^{93}	>100
Ru	4.4-hr Ru^{105}	>20
Ba	38.8-hr Ba^{133m}	>100
Tl	1.8-hr Tl^{198}+7-hr Tl^{199}	200
Pb	~80-min Pb^{199}	8

II. EXPERIMENTAL PROCEDURE

Mercury was irradiated with 122-Mev π^- mesons. The mesons were produced by the bombardment of a beryllium target with 450-Mev protons, were deflected by the fringing field of the cyclotron magnet through a 6-in. channel in the iron-concrete cyclotron shield (6- to 12-ft thick), and were then deflected by an auxiliary magnet through an angle of about 45 deg. The mesons leaving the magnet were in a beam about 4 in. on edge. The intensity of the meson beam was measured by coincidence counting of the pulses of two 4-in. $\times$6-in.$\times\frac{1}{2}$-in. thick scintillation counters, placed about 4 ft from the magnet.[13] In the various experimental arrangements, the meson counting rates varied between 40 000 and 100 000 mesons/min for a proton beam dissipating 1 watt of power in the beryllium target.

The mercury irradiations were performed at meson intensities ranging from 4×10^5 to 3×10^6 mesons/min, and for irradiation times of 2 to 8 hr. The mercury was contained in a 5-in. square vessel about $2\frac{1}{2}$ in. deep. Generally, about 10 kg of mercury was used. After the irradiation, the mercury was transferred to a large sintered glass funnel. A wash solution consisting of $0.1M$ HNO_3 and carriers of the elements to be isolated was added above the mercury, and the mercury was agitated by passing air through it for about 20 minutes in order to extract the desired radioactive species. The wash solution was withdrawn and radiochemical analyses were performed.[14]

Experiments were done to test the efficiency of extraction of the radioactive species from the mercury to the wash solution and the retention of radioactivity on the walls of the vessel. Mercury was irradiated with

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[1] A. Turkevich and J. B. Niday, Phys. Rev. **84**, 1253 (1951).

[2] T. T. Sugihara and W. F. Libby, Phys. Rev. **88**, 587 (1952).

[3] A. Turkevich and J. B. Niday, Phys. Rev. **90**, 342 (1953).

[4] S. C. Fung and A. Turkevich, Phys. Rev. **91**, 480 (1953).

[5] L. Winsberg, Phys. Rev. **90**, 343 (1953).

[6] N. Sugarman, Phys. Rev. **86**, 604 (1952); N. Sugarman and A. Haber, Phys. Rev. **90**, 342 (1953).

[7] S. G. Al-Salam, Phys. Rev. **84**, 254 (1951).

[8] W. John and W. F. Fry, Phys. Rev. **91**, 1234 (1953).

[9] R. H. Goekermann and I. Perlman, Phys. Rev. **76**, 628 (1949).

[10] P. R. O'Connor and G. T. Seaborg, Phys. Rev. **74**, 1189 (1948).

[11] L. Jodra and P. Kruger, University of Chicago, unpublished work.

[12] S. Tamor, Phys. Rev. **77**, 412 (1950).

[13] The experimental arrangement does not differ significantly from that given in Fig. 1, reference 1.

[14] *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Part VI, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.

neutrons outside the cyclotron tank. The radioactivity in the sample was many times that obtained in the meson irradiations. The mercury was subjected to the washing process, the wash solution was withdrawn, and the mercury was washed again. The ratio of the activity of a given nuclide in the first wash solution to that in the second measures the efficiency of removal of that element from the mercury, if equilibrium is established. The data are given in Table I. There is a large range in washing ratios, varying from 5 for the less electropositive elements (arsenic and lead) to greater than 100 for the more electropositive elements (strontium, yttrium, barium, thallium). A low washing ratio indicates that a substantial fraction of the radioactivity remains in the mercury. It is assumed that interchange occurs between the carrier and tracer, such that the chemical yield correction for the carrier takes account of the low washing ratio. In a separate experiment it was found that the activity retained on the walls amounted to about 1 percent of the total activity.

The radioactivity measurements were made with end-window methane-flow proportional counters, operating at about 4000 volts. The counter window is made of rubber hydrochloride, $\sim$0.7 mg/cm^2, Aquadaged on both sides. The backgrounds of the counters are about 9 counts/min, and are reproducible to about 0.3 count/min. The counting was usually reliable even for counting rates as low as 1 count/min above background. The isolated samples had counting rates ranging from 1 to 150 counts/min depending upon the half-life, meson intensity, and irradiation time. The samples were positioned about 1.5 mm from the counter window, and the geometry factor as determined with a RaDEF standard was found to be 34 percent for weightless samples with no backing, and 47 percent for samples of about 10 mg/cm^2, mounted on $\frac{1}{16}$-in. aluminum cards.

Experiments were also performed on the fission of mercury with high-energy neutrons. The fissionability of mercury by neutrons of energy up to 90 Mev had been reported earlier.[15] The neutrons are produced from protons on beryllium and have an energy distribution from 200 to 440 Mev, with a peak at about 350 Mev.[16,17] The neutron flux is about 2×10^5 neutrons/cm^2 sec over an area of about 50 cm^2. The activity from the neutron-irradiated samples was about one hundred times that from the meson-irradiated samples, which made it possible to identify some of the long-lived components, whereas this was not possible in the meson experiments.

III. RESULTS

A. Fission

The elements isolated from the forty-meson and ten-neutron irradiations include some fission products as well as thallium and lead. Since the observed activity in a given sample was about 50 counts/min or less for the meson irradiations, it was assumed to be composed of known nuclides, and the decay curves were analyzed accordingly. The activity of an individual nuclide was corrected for chemcial yield and time of irradiation. The ratio of the saturation activity of the nuclide to that of the "7-hr" Sr (9.7-hr Sr^{91}, 2.7-hr Sr^{92} and 3.5-hr Y^{92} decaying with a half-life of 7 to 8 hr) was obtained as a measure of the yield of the nuclide. The data are given in Table II. It was found that the ratios thus obtained did not vary outside the experimental errors, estimated at about 20 percent for the meson data and about 10 percent for the neutron data, when the irradiation time was varied, despite the complexity of the species of which the "7-hr" Sr activity is composed. The ratio of Sr^{89} to "7-hr" Sr of $\sim$1.5 for the meson experiments was the result of about twenty experiments in each of which the counting rate of Sr^{89} was about 0.5 count/min above background.

The fission yields of Table II were calculated on the assumption that Sr^{89} has a fission yield of 5.0 percent. This is the yield expected if the yield-mass distribution for the fission of mercury with mesons or high energy neutrons is single-humped with Sr^{89} near the maximum, similar to that of bismuth with high-energy deuterons or protons.[9,11] The general trend of the yield data, the relatively high yield of Br^{82}, and the absence of barium activity are all consistent with a high-energy fission process.

TABLE II. Activity data of fission products from meson- and neutron-irradiated mercury.

	Ratio of activity to "7-hr" Sr		Fission yield, percent	
Nuclide	Meson	Neutron	Meson	Neutron
17.5-day As^{74}		0.12		0.4
26-hr As^{76}		0.35		1.2
38-hr As^{77}		0.59		2.0
91-min As^{78}	$\sim$0.3	0.51	$\sim$1.0	1.7
4.4-hr Br^{80m}		1.0		3.3
36-hr Br^{82}	$\sim$0.3	0.41	$\sim$1.0	1.4
2.4-hr Br^{83}	0.81	0.45	2.7	1.5
19.5-day Rb^{86}	<3	1.3	<10	4.3
54-day Sr^{89}	$\sim$1.5	1.5	(5.0)	(5.0)
9.7-hr Sr^{91}		0.64		2.1
2.7-hr Sr^{92}		0.18		0.60
60-hr Y^{90}		1.4[a]		4.7[a]
3.5-hr Y^{92}	2.0[b]	1.4[b]	6.7[b]	4.7[b]
10-hr Y^{93}	1.7	1.2	5.7	4.0
4.4-hr Ru^{105}	0.47	0.47	1.6	1.6
2.0-hr Ba^{129}	<0.03	0.009	<0.1	0.03
38.8-hr Ba^{133m}	<0.2[c]	0.041[c]	<0.67[c]	0.14[c]
12.0-day Ba^{131} +12.8-day Ba^{140}		0.01[c]		0.033[c]
85-min Ba^{139}	<0.03		<0.1	

[a] Independent yield of Y^{90}.

[b] Calculated as being produced directly in fission, independent of the Sr^{92} parent.

[c] Results uncorrected for counting efficiency of nucilde.

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[15] E. L. Kelly and C. Wiegand, Phys. Rev. 73, 1135 (1948).

[16] J. Marshall and V. A. Nedzel, University of Chicago, unpublished work.

[17] Goodell, Loar, Durbin, and Havens, Phys. Rev. 89, 724 (1953).

TABLE III. Activity data of thallium and lead nuclides from meson-, neutron-, and proton-irradiated mercury.

Nuclide	Half-life, reported	Half-life, observed	Ratio of activity to "7-hr" Sr, uncorrected	Counting corr. factor	Ratio to "7-hr" Sr, corr.	Branching ratio		
				Meson		Tl atoms/meson		
Tl^{198}	1.8 hr	1.8–2 hr	2.3	3.5	8.0	1.57×10^{-3}		
Tl^{199}	7 hr	7–8 hr	0.83	8.8	7.3	1.43×10^{-3}		
Pb^{199}	~80 min		<0.025					
Pb^{201}	8 hr		<0.03					
				Neutron				
Tl^{198}	1.8 hr	1.7–1.8 hr	5.6	3.5	19.6			
Tl^{199}	7 hr	6–7 hr	1.6	8.8	14.1			
Tl^{200}	27 hr	22–25 hr	0.48	~13	~6			
Tl^{201}	72 hr	64–67 hr	0.31	~19	~6			
Pb^{199}	~80 min	95 min	0.03					
Pb^{201}	8 hr	10 hr	0.01					
				Proton		Tl atoms/proton		
						37 Mev	55 Mev	72 Mev
Tl^{198}	1.8 hr	1.7–1.9 hr		3.5		3.7×10^{-3}	1.15×10^{-2}	1.4×10^{-2}
Tl^{199}	7 hr	6.7–7 hr		8.8		6.0×10^{-3}	1.05×10^{-2}	9.6×10^{-3}

In general, the yields of the fission products from meson fission agree with those high-energy neutron fission. The difference in the ratio of Br^{83} to Br^{82} in meson fission, as compared to neutron fission, is probably associated with the fact that meson fission is a lower energy process than neutron fission. The ratios of Br^{80m} to Br^{82} and Br^{83} to Br^{82} for the neutron fission of mercury are the same as those found in the fission of gold with 450 Mev protons,[11] whereas in the meson experiments, Br^{80m} was not evident and Br^{83} was correspondingly in much higher yield. Experiments on bismuth with varying proton energy[11] demonstrated that the ratios Br^{83} to Br^{82} and Br^{80m} to Br^{82} are energy dependent, the former increases and the latter decreases as the proton energy is lowered. Since these ratios do not change rapidly with the atomic number of the target nucleus, a comparison of the Br^{83} to Br^{82} ratio in meson fission of mercury with that in proton fission of bismuth leads to the conclusion that the meson fission process corresponds roughly to fission occurring with protons of about 150 Mev.

Experiments were performed to determine the fission-product activity induced in mercury by the stray radiation in the vicinity of the sample. The cyclotron was operated at the usual intensity of about 20 watts on the beryllium target, and the sample was irradiated at its usual position relative to the deflecting magnet. The current in the deflecting magnet was turned off. Under these circumstances, the counting rate in the scintillation counters used in measuring meson intensity was about 0.2 percent of that obtained when the magnet current was at the optimal value. Strontium analyses were performed on the mercury and it was found that the activity was low, about 5 percent of that produced in the meson irradiations.

The possibility that the observed fission of mercury was not the primary effect of meson interaction, but rather the effect of secondary interaction of fast neutrons and protons produced by meson interaction, was tested by irradiating a mercury sample directly behind another whose thickness was slightly greater than the range of the mesons in mercury. No activity of strontium was observed other than that expected from stray background radiation. This result is consistent with that calculated for secondary fission on the assumption that as many as one neutron or proton of about 60-Mev energy is produced for each meson interaction. The calculated fission contribution from the secondary particles is less than 1 percent of the observed fission under the usual irradiation conditions.

B. Thallium Production

Thallium was separated[18] from the meson-irradiated mercury and activity of about 100 counts/min was found which could be resolved into two components, 1.8-hr Tl^{198} and 7-hr Tl^{199}. There was also evidence for a longer-lived species, but the counting rate was less than 1 count/min, so no assignment of the activity could be made. About 5 percent of the thallium activity was produced by stray radiation, as determined in experiments in which the deflecting magnet was off. Thallium activity was also found in the neutron-irradiated mercury, and in this case the activity was high enough so that the decay curves could be analyzed into the periods reported[19] for masses 198 to 201. The yields of the thallium isotopes relative to the yield of "7-hr" Sr from fission, uncorrected for the counting efficiencies of these electron-capture species, are given in column 4 of Table III. A serach for lead activity in meson-irradiated mercury led only to the upper limits for the yields of Pb^{199} and Pb^{201} given in column 4 of Table III. Lead

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[18] The chemical procedures for the separation of thallium and lead were those given by W. W. Meinke, University of California Radiation Laboratory Report UCRL-432, 1949 (unpublished), with some modifications.

[19] Hollander, Perlman, and Seaborg, *Table of Isotopes*, Revs. Modern Phys. **25**, 469 (1953).

activity was observed in the neutron-irradiated mercury, which could be attributed to Pb^{199} and Pb^{201}. The yields of these nuclides are given in Table III.

The thallium yield data of column 6 of Table III were obtained from those of column 4 by correcting for the counting efficiencies of thallium isotopes. Counting correction factors (column 5) were found by determining the counting rates of the K x-rays of the thallium isotopes in NaI crystal counters[20] and by comparing these counting rates, corrected for fluorescence yield and absorption, with those of the conversion electrons in the methane proportional counter. It was assumed in this work that all of the thallium isotopes decay by electron-capture processes.[21]

In order to determine the branching ratio of Tl^{198} absolutely, two experiments were performed in which the meson intensity was carefully measured and precautions were taken against loss of mesons from scattering out of the sample. The mesons were collimated by a 6-in. thick iron collimator with a 1-in. aperture. The sample, contained in the 5-in. square vessel, was placed at the position occupied by the counters, about 1 ft from the collimator. The collimator reduced the dimensions of the meson beam so that the scattered mesons would not leave the mercury sample. The meson intensity was reduced about sevenfold, and about 10 percent of the counting rate in the scintillation counters was attributed to energetic electrons and μ mesons. The initial counting rate of the thallium samples was about 20 counts/min. The ratio of the saturation activity of Tl^{198}, corrected for the geometry factor of counting but uncorrected for the counting efficiency of the radiations, to the meson intensity was found to be 4.5×10^{-4}. Using the value of the counting correction factor of 3.5, this ratio becomes 1.57×10^{-3} Tl^{198} atoms/meson. The meson branching ratio for Tl^{199} can be calculated from the data of column 6 and the branching ratio for Tl^{198} and is found to be 1.43×10^{-3} Tl^{199} atoms/meson. These results are presented in column 7, Table III.

C. Branching Ratio for Fission

The branching ratio for fission can, in principle, be determined by isolation of a suitable fission product of known fission yield from the meson-irradiated mercury used for the Tl^{198} branching ratio determination. Strontium was isolated in these two experiments but the activity was only about 5 counts/min. The data of Tables II and III on the yield of Tl^{198} relative to "7-hr" Sr and the branching ratio of Tl^{198} may be used to calculate the branching ratio for fission. If the fission yield of Sr^{89} is assumed to be 5 percent, the branching ratio for fission is calculated to be 5.9×10^{-3}. Since all of the pions interact with mercury, either during the slowing down process from 120 Mev to rest, or after capture into an atomic orbital, it is evident that the fraction of these interactions giving rise to fission is about 1 in 200.

IV. DISCUSSION

A. Fission Branching Ratio

The low branching ratio of about 0.5 percent for fission of mercury by pions can be understood on the basis of the current models for the interaction of pions with nuclei and the fission cross section of mercury with high energy particles. If it is assumed that the meson interacts with two nucleons and shares its energy between them,[12] then the excitation given the nucleus is essentially the same as that given it by a high-energy proton or neutron whose kinetic energy is equal to the sum of the kinetic and rest-mass energy of the meson. For mesons of low kinetic energy, where multiple particle interaction is more likely, comparison with fission by deuterons of the same total energy is perhaps more valid. The expected branching ratio for fission from these considerations would be the ratio of the fission cross section to the total cross section with high-energy protons and deuterons. For gold this ratio is about 1 percent.[22] It is expected, from the results of the fission experiments done with neutrons of up to 90 Mev energy,[15] that the cross section for fission of mercury would be only slightly higher than that of gold. It is thus evident that the fission branching ratio of mesons is in fair agreement with that expected from the model of the meson interaction with the nucleus.

B. Tl^{198} Branching Ratio

The production of thallium activity in the mercury may be explained by the interaction of the fast protons expected from the meson interactions with mercury. It is found in photographic plate studies that 0.3 fast proton of about 55 Mev is emitted for each fast meson interaction[23,24] (30 to 90 Mev), and 0.1 fast proton for each slow meson.[25] Since about 40 percent of the 122-Mev π^- mesons would interact in flight while being slowed down, assuming a cross section of 2 barns as observed for 85–137 Mev pions,[26] and 60 percent at rest, then the number of fast protons expected per meson is 0.18. On

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[20] The scintillation counting of the thallium samples was done by Dr. J. R. Arnold of the Institute for Nuclear Studies of the University of Chicago and by Dr. D. W. Engelkemeir of the Argonne National Laboratory. The correction factors determined independently agreed to about 20 percent.

[21] Recent work of I. Bergström and R. D. Hill [Bull. Am. Phys. Soc. **28**, No. 4, 13 (1953)] shows that there is a 2-hr period among the thallium isotopes decaying by isomeric transition, and that the ground state of Tl^{198} has a half-life of $\sim$4 hr. These results affects the yields reported here, but do not appreciably affect comparisons of production of a given thallium isotope by mesons, neutrons, or protons.

[22] J. Jungerman, Phys. Rev. **79**, 632 (1950).

[23] Bernardini, Booth, Lederman, and Tinlot, Phys. Rev. **82**, 105 (1951).

[24] G. Bernardini, Proc. Inter. Conf. Nuc. Phys. and Phys. Fundamental Particles, Univeristy of Chicago, September 17, 1951, p. 13 (unpublished).

[25] F. L. Adelman, Phys. Rev. **85**, 249 (1952).

[26] R. L. Martin, Phys. Rev. **87**, 1052 (1952).

the assumption that the number and energy of fast protons from meson interactions in mercury is the same as that found in photographic plates,[27] it is possible to calculate the expected production of Tl^{198} per meson. The reaction range of a 55 Mev proton in mercury[28] is 5.3 g/cm^2, after correction for the Coulomb barrier. If it is assumed that the only reactions which occur in appreciable yield are reactions in which neutrons are emitted, such as $(p,5n)$, $(p,4n)$, etc., and that each of these reactions occurs in succession, as the proton is being slowed down, with a geometric cross section of 2 barns and an average isotopic abundance of 0.19, then the expected yield of Tl^{198} per proton is 6.1×10^{-3}. The yield per meson is 1.1×10^{-3} for 0.18 fast protons per meson. This value compares favorably with that observed experimentally of 1.57×10^{-3}.

In view of the numerous assumptions needed to understand the production of thallium by secondary fast protons from mesons, some experiments were performed to measure directly the production of thallium activity from protons of 37 to 72 Mev. This information then permits a direct comparison with the thallium activity produced by mesons without assumptions on cross sections for (p,xn) reactions. The low-energy protons were produced by backward scattering from the meson target and deflection into the 122-Mev π channel of the 12-ft shield by the fringing field of the cyclotron magnet.[29] Further deflection by the deflecting magnet produced beams of protons of well-defined energy in good intensity, where the energy was controlled by the target position and the deflecting magnet current. Absorption measurements were made on three proton beams using two 2-in. diameter scintillation counters in coincidence. From the ranges, the proton energies[28] were found to be 40±3 Mev, 57±4 Mev, and 74±4 Mev. The intensity in the three beams was about 40 000 protons/min for a cyclotron beam intensity of 1 watt. Proton irradiations were made with the three beams with the cyclotron operating at about 20 watts, using a mercury vessel 3 in. square in cross section and 1 cm deep. The proton energies were reduced somewhat by the $\frac{1}{32}$-in. aluminum wall of the irradiation vessel. Thallium was isolated from the proton-irradiated mercury and the decay curves were resolved into Tl^{198}, Tl^{199}, and a long-lived component, probably a mixture of Tl^{200} and Tl^{201}. Strontium samples isolated from the mercury were inactive, in keeping with the low cross section for fission at these energies.[22]

The yields of Tl^{198} and Tl^{199} for the proton-irradiated mercury are given in column 7 of Table III. It is seen that the yield of Tl^{198} per 55-Mev proton of 1.15×10^{-2} agrees well with that estimated earlier of 6.1×10^{-3}. The variation of the yields with energy can be used to estimate the average proton energy in the meson experiments. The ratio, Tl^{198}/Tl^{199}, in the meson experiments is about the same as that found for 55-Mev protons, the average energy of fast protons from mesons in emulsion. Assuming, then, that the average proton energy from meson interaction in mercury is about 55 Mev, we find that the number of such protons per meson is about 0.14, again in good agreement with that found in emulsion studies, 0.18. It is evident, then, that secondary proton bombardment satisfactorily explains the production of thallium in the meson-irradiated mercury. Similarly, the production of thallium and lead activities in the neutron-activitated mercury may be explained by secondary proton and alpha particle bombardment. The ratio Tl^{198}/Tl^{199} from the neutron-activated mercury is about that for protons of about 70 Mev.

It is a pleasure to acknowledge the advice of Professor E. Fermi and Professor A. Turkevich, and the cooperation of Professor H. L. Anderson, Mr. L. Kornblith, and members of the synchrocyclotron crew in these experiments.

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[27] Preliminary observations of fast protons produced from meson interactions with mercury have been made by E. Silverstein and J. Orear of the Institute for Nuclear Studies, University of Chicago, using photographic plate techniques. Fast protons were observed although there are as yet no quantitative data on their yield.

[28] W. A. Aron, University of California Radiation Laboratory Report UCRL-1325, May 1951 (unpublished).

[29] The production of collimated, monoenergetic, low-energy proton beams by this technique was worked out by E. Fermi early in 1952. The experimental arrangement is essentially the same as that used in the meson irradiations.