$$\sum_k r_{jk}^{-6} = 8.4\ d^{-6}, \qquad (A.17)$$

which leads to (9). Assuming that the exchange interaction is isotropic and of the form

$$A(r_{jk}) \begin{cases} = A & r_{jk} < d \\ = 0 & r_{jk} > d \end{cases}$$

for a simple cubic lattice and $s = 1/2$, as in reference 4, Eq. (A.6) is easily transformed to the expression

$$\omega_{20} = 3.65\,|A|/\hbar. \qquad (A.18)$$

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SOVIET PHYSICS JETP VOLUME 35 (8), NUMBER 1 JANUARY, 1959

*SECONDARY NUCLEAR REACTIONS INDUCED IN BISMUTH AND LEAD BY HIGH-ENERGY PROTON BOMBARDMENT**

B. V. KURCHATOV, V. N. MEKHEDOV, L. V. CHISTIAKOV, M. Ia. KUZNETSOVA, N. I. BORISOVA, and V. G. SOLOV' EV

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The production of astatine isotopes ($Z = 85$) by bombardment of bismuth ($Z = 83$) and lead ($Z = 82$) with protons of energies up to 480 Mev was studied radiochemically. The cross sections for At^{211} and At^{210} production from bismuth were found to be 6×10^{-29} and $\sim 2 \times 10^{-29}$ cm^2 respectively. The formation of the light isotopes At^{205} and At^{203} was established. At^{211} was detected in lead ($\sigma \sim 10^{-31}$ cm^2).

The experimental results are attributed to secondary capture of the disintegration products (α particles or lithium nuclei). The production of light astatine isotopes is explained by high-energy proton capture with subsequent emission of π^- mesons and several neutrons. The cross section for production of α particles with $E > 20$ Mev from bismuth irradiated by 480-Mev protons was determined from the astatine yield to be 5 to 6×10^{-25} cm^2.

ONE of the characteristics of interactions between complex nuclei and fast protons is the occurrence of reactions in which the charge of the target nucleus is increased by 2 or 3. Such reactions have frequently been described in the literature[1-3] and are attributed to secondary capture of disintegration products (α particles or lithium nuclei). At high proton energies, in addition to such so-called secondary reactions, there is the additional possibility of proton capture accompanied by the emission of a π^- meson and neutrons, thus also increasing the charge by 2.

*The experimental work on which this paper is based was performed in 1951 and 1952 (cf. Report of the Institute of Nuclear Problems, Academy of Sciences, U.S.S.R. for 1951).

In the present paper we present the results of an investigation of the indicated reactions in bismuth and lead under bombardment by fast protons with energies up to 480 Mev. The production of nuclides with Z increased by 2 or more is clearly revealed through the appearance of α-radioactive astatine isotopes. Investigation of the yields of different astatine isotopes revealed both mechanisms of charge increase and enabled us to explore certain quantitative aspects of the effect which had not been studied previously.

THE PRODUCTION OF ASTATINE FROM BISMUTH

The initial material for the experiments was bismuth of high purity, shown by chemical analysis to contain < 0.001% of thorium and < 0.01% of uranium. The metal plates used in the experiments measured 10×15×2 mm and weighed up to 2 g. The plates were wrapped in aluminum foil 25 μ thick and were bombarded by a 0.2 to 0.4 μa proton current. Different proton energies from 180 to 480 Mev were obtained by varying the radius of the target position in the accelerator. The geometry of the bombardment was such that the beam passed through each plate in the direction of the 10-mm side. The initial energy of the incident proton beam was varied from 5 to 17% while the period of the bombardment ranged from 7 to 20 min.

The aluminum foil was used to monitor the current. After bombardment, the foil was unwrapped and the amount of Na^{24} activity with the half-life T = 14.8 hrs was measured. The cross section for Na^{24} production from Al^{27} was assumed to be 10^{-26} cm^2.[3]

Chemistry of Separation. The astatine was separated through coprecipitation of astatine and tellurium and the distillation of atomic astatine from an aqueous solution.[4,5] The astatine was extracted by different methods, depending on the purity and time requirements.

Quantitative Separation of Astatine. A bombarded specimen was dissolved in a minimum amount of concentrated nitric acid. The solution was diluted by 10% hydrochloric acid and an aliquot part of about 1/10 was combined with dilute hydrochloric acid to a volume of 30 to 40 milliliters. After the addition of 50 mg of tellurium (0.25 ml of a 2% solution of tellurium in nitric acid) tellurium was precipitated by 1 N stannous chloride. The precipitate was separated by centrifuging. Since a single precipitation of astatine in tellurium is insufficient, the operation was performed three times. The tellurium precipitates were combined and washed four times in 25 to 30 ml of dilute hydrochloric acid. (Such careful washing is essential for complete removal of the bismuth. If the washing is inadequate, the $Bi(OH)_3$ deposited by alkalization during the operations retains a large fraction of the astatine.) The washed tellurium was dissolved in a few drops of concentrated nitric acid; the solution was then diluted with water to a volume of 10 to 15 ml and alkalized by a 20% solution of sodium hydroxide. The tellurium was then precipitated by adding a stannite for the purpose of removing the polonium from the solution. Under these conditions, according to the literature,[4] the astatine remains in the solution. The purification from polonium was repeated. The purified solution was acidified with concentrated hydrochloric acid, 2 to 3 mg of tellurium was added, and the tellurium was precipitated by stannous chloride. The precipitation was repeated twice. The deposits were washed in water, deposited with alcohol on stainless steel plates (d = 2.5 cm), and dried on a water bath, after which the α activity was determined. The coprecipitation of astatine and tellurium is illustrated by the values given below, which are the averages of seven experiments. The amount of the carrier before the first and second precipitations was 2 mg, while the amount was 3 mg before the third precipitation. In the first precipitation 80% of the astatine was extracted, in the second precipitation 17.6%, and in the third precipitation 2.4%.

Preparation of Pure Astatine. The astatine was precipitated with tellurium by a similar method out of the solution which remained after removal of the aliquot part. The precipitates were combined and washed, and the tellurium was dissolved in 2 ml of concentrated sulfuric acid to which two drops of concentrated nitric acid had been added. The solution was diluted with water to 80 to 100 ml, 2 g of $FeSO_4$ were added, and the astatine was distilled, with 10 ml of the solution remaining in the distilling flask. The distillate was received in a vessel containing a solution of sodium hydroxide; 5 mg of tellurium was added and the latter was precipitated by adding a stannite for purification from the residue of polonium and β and γ emitters. In order to determine the amount of distilled astatine, an aliquot part of the alkaline solution was taken and the astatine was separated by the method described above. After 3 or 4 days, the daughter Po^{210} was precipitated with tellurium from the remaining solution, using the stannite in an alkaline medium, to determine the yield of At^{210}.

Rapid Separation of Pure Astatine. The sample was dissolved in concentrated nitric acid and the excess of acid was neutralized by a sodium hydroxide solution to a degree sufficient to keep the bismuth dissolved during the experiment. The solution was diluted with hot water to a volume of 10 ml and was boiled for 5 min, with the astatine being collected by the alkaline solution. About 1/20 of the astatine was boiled off with the admixture of β and γ emitters. Tellurium was precipitated from the distillate after the addition of tellurium and a stannite solution. Acidification and the addition of tellurium in the solution thus purified caused the astative to precipitate and be deposited on the target, as stated above. The entire operation of separation and preparation of the target required 20 to 25 min. This portion of the astatine was intended for the detection and measurement of short-lived α-radioactive isotopes and the determination of their relative yields.

PROCEDURE AND EXPERIMENTAL RESULTS

The α activity of the astatine targets was measured by means of an ionization chamber in conjunction with a linear accelerator. Some of the targets were also measured with a Geiger counter to study the astatine isotopes which decay through the capture of an orbital electron.

In all of the experiments we observed α activity with a half-life from 7.0 to 7.8 hrs, corresponding to At^{211} (α, K 60%), T = 7.5 hrs. The counter measurements revealed an eight-hour half-life belonging to At^{210}(K), T = 8.3 hrs. The production of the latter isotope was also confirmed by the accumulation of the long-lived α activity of the daughter Po^{210} (T = 140 days).

The utilization of the very rapid method of separating pure astatine enabled us to detect shorter periods of α activity 25 to 30 min and about 5 min, ascribed to At^{205}(α, K), (T = 25 min) and At^{203}(α, K), (T = 7 min) respectively.[6] The activities of At^{209}(α, K 95%) (T = 5.5 hrs) and At^{207}(α, K 90%) (T = 2 hrs) could not be separated on the decay curve because of the closeness of the half-lives and the small relative amount of α emission. The isotopes At^{204}(K), (T = 25 min), At^{206}(K), (T = 2.5 hrs), and At^{208}(K), (T = 6.3 hrs), which decay only by capture of an orbital electron, were not investigated.

The activity of At^{211} per gram of bismuth, under continuous bombardment and taking into account the percentage of K capture, fluctuated from 2.1×10^7 to 4.5×10^7 decays per min. The yields could not be determined with high accuracy because of a number of methodological errors. In addition to sorption losses during chemical separation, there were possible errors resulting from variations of the size of samples and the geometry of irradiation. We could also not exclude the possible loss of a small amount of astatine by evaporation from the bismuth during the dissimilar heating of the samples under bombardment.

The results obtained through bombardment by 480-Mev and 180-Mev protons are given in Tables I and II. Comparison of the average values shows

TABLE I*

Weight of Bi, g	Weight of Al, mg	A_∞ (At^{211}) per gram-atom of Bi, decays/min	A_∞ (Na^{24}) per gram-atom of Al, decays/min	σ (At^{211}), cm²	$\frac{\sigma(At^{210})}{\sigma(At^{211})}$
1.95	32.4	$4.3 \cdot 10^9$	$5.3 \cdot 10^{11}$	$7.4 \cdot 10^{-29}$	0.43
1.69	25.5	$2.9 \cdot 10^9$	$9.1 \cdot 10^{11}$	$5.4 \cdot 10^{-29}$	0.28
1.25	18.5	$9.3 \cdot 10^9$	$1.6 \cdot 10^{12}$	$5.8 \cdot 10^{-29}$	0.31
1.18	22.5	$8.7 \cdot 10^9$	$1.5 \cdot 10^{12}$	$5.8 \cdot 10^{-29}$	0.27
0.7	2.2	$1.4 \cdot 10^{10}$	$1.9 \cdot 10^{12}$	$7.4 \cdot 10^{-29}$	
	Average			$6.3 \cdot 10^{-29}$	0.32

*E_p = 480 Mev.

TABLE II*

Weight of Bi, g	Weight of Al, mg	A_∞ (At^{211}) per gram-atom of Bi, decays/min	A_∞ (Na^{24}) per gram-atom of Al, decays/min	σ (At^{211}), cm²	$\frac{\sigma(At^{210})}{\sigma(At^{211})}$
1.9	21.8	$3.6 \cdot 10^9$	$1.7 \cdot 10^{12}$	$2.1 \cdot 10^{-29}$	0.33
1.64	26.8	$8.7 \cdot 10^8$	$2.3 \cdot 10^{11}$	$3.8 \cdot 10^{-29}$	0.34
1.16	21.5	$3.5 \cdot 10^9$	$9.7 \cdot 10^{11}$	$3.6 \cdot 10^{-29}$	0.28
			Average	$3.2 \cdot 10^{-29}$	0.32

*E_p = 180 Mev.

TABLE III

E_p, Mev	$\sigma(At^{205})/\sigma(At^{211})$	$\sigma(At^{203})/\sigma(At^{211})$
480	0.035	~0.25
375	0.07	~0.5
275	0.12	—
180	<0.02	<0.2

that the yield of At^{211} is nearly twice as great for 480-Mev protons as for 180-Mev protons, whereas the ratio of the At^{210} and At^{211} production cross sections is unchanged. Table III shows how the cross section ratios vary for At^{205} and At^{211} and for At^{203} and At^{211} as the proton energy decreases.

The yields of At^{205} and At^{203} are lower limits because we do not know for these isotopes the relative amount of K capture which competes with α decay. The decay curve does not show a five-minute period for 275-Mev protons, and the 25-minute period disappears for $E_p = 180$ Mev. The values given in Table III are only estimates; greater accuracy will require improved apparatus, especially a multi-channel analyzer of α-particle energies.

DISCUSSION OF RESULTS

The production of the heavy isotope At^{211} cannot be attributed to the disintegration of uranium and thorium impurities. Control experiments for the purpose of determining the At^{211} yield from these elements gave cross sections $\sigma = 2.3 \times 10^{-26}$ cm^2 for thorium and 3.0×10^{-28} cm^2 for uranium. Hence the observed effect would require ~0.3% of thorium or ~10% of uranium, which considerably exceeds the limit of impurities represented by these elements.

The only reaction which accounts both for the increased charge and the mass of the products is the capture of α particles with emission of two or three neutrons. Since an admixture of α particles in the proton beam is unlikely, we assume a secondary origin of the α particles resulting from the disintegration of target atoms. We are able to relate the yields of At^{210} and At^{211} quantitatively to the observed α-particle spectrum of bismuth and to obtain the correct order of magnitude of the cross section for the production of fast α particles. The results of these calculations, and a detailed discussion of all questions arising therefrom, are given separately in the following section.

The cross sections obtained for secondary α-particle capture by bismuth are in agreement with the cross sections of similar secondary reactions in copper.[2] Without taking into account the different energies of the bombarding particles, it can be assumed that secondary capture of α particles occurs with a cross section of the same order of magnitude ($\sim 5 \times 10^{-29}$ cm^2) for various elements of the periodic system.

The foregoing mechanism for the production of heavy astatine isotopes cannot be extended to include the light isotopes. Indeed, we know from the literature that the production of At^{205} and At^{204} from bismuth requires 150-Mev α particles, while At^{203} is obtained through bombardment at 275 Mev.[6] Aside from the difficulty of explaining a considerable yield for the emission of such fast α particles, any appreciable quantity of these α particles would result in the predominance of At^{210} over At^{211} (owing to the large total (α, 3n) cross section; see Fig. 1), in contradiction of experiment. Therefore the production of light isotopes with a yield of the same order or one order smaller than the yield of At^{211} cannot be accounted for by secondary reactions with α particles, and requires a mechanism of π^--meson emission. The corresponding reaction for At^{203} production can be written as $Bi^{209}(p, \pi^- 7n)At^{203}$. The integral π^--emission cross section for heavy nuclei is $\sim 10^{-26}$ cm^2.[7] The cross section of the assumed particular reaction must be smaller by 2 to 3 orders of magnitude, thus not disagreeing with the observed yield of light astatine isotopes. The reaction mentioned necessitates the capture of a fast proton and the transfer of its total energy to the nucleus.

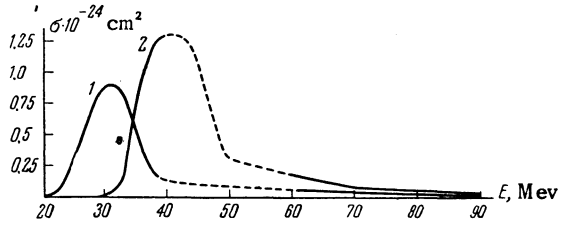

FIG. 1. Excitation functions for the (α, 2n) and (α, 3n) reactions in bismuth. 1 – Bi(α, 2n)At^{211}; 2 – Bi(α, 3n)At^{210}.

Taking into account the energy required for the production of a π^- meson (170 Mev) together with its kinetic energy (up to 100 or 200 Mev), we obtain the nuclear excitation energy remaining for nucleon evaporation; this is about 100 to 200 Mev. The high excitation energy will favor the emission of a large number of nucleons (in our case of production of astatine from bismuth, these can only be neutrons); thus the production of the lightest isotopes will be favored. This can also explain our observation of the predominance of

At^{203} over At^{205} and the increased yield of At^{205} as the incident proton energy decreases.

Calculation of the Yield. Figure 1 shows the excitation functions for the (α, 2n) and (α, 3n) reactions in bismuth;[6,8] the dotted lines denote our interpolations of missing portions of the curves. These experimental results furnish the only possibility to date of a quantitative analysis of a secondary reaction involving α-particle capture.

The calculation was performed as follows:

1. It was assumed that heavy astatine isotopes were produced through the capture of α particles with $20 < E < 60$ Mev emitted by bismuth when bombarded by fast protons.

2. It was necessary to determine a shape of the α-particle spectrum which would satisfy the observed ratio of At^{210} and At^{211} yields. After trials of the simplest functions that satisfy experiment (linear decay and a steep E^{-n} law) we took an intermediate spectral shape given by the theory of evaporation[9] as expressive of the physical content:

$$P(E) = \frac{E-V}{\tau^2} \exp\{-(E-V)/\tau\}.$$

3. We tried different values of the parameters V and τ, which in evaporation theory are the potential barrier and temperature of the nucleus.

4. For each pair of values of V and τ, we determined graphically the fraction K_n of α particles with $E < 20$ Mev at 5-Mev intervals, so that

$$\sum_{E=20}^{E=60} K_n = 1.$$

5. For each group of α particles, we determined the probability of α-particle capture with emission of two and three neutrons respectively. This probability W_n was obtained by summation over the entire effective range of probabilities for different portions Δl of the α-particle range. The probability w was calculated by means of the formula

$$w = \Delta l/\lambda = \Delta l n_0 \sigma_{At},$$

where n_0 is the number of Bi atoms per cm^3 and σ_{At} is the cross section for the production of the corresponding astatine isotope for a given energy interval. Δl was determined from the graph of the energy dependence of the α-particle range.[10]

6. Summation of the products $K_n W_n$ from 20 to 60 Mev gave the total probability of α-particle capture with formation of the corresponding isotope.

7. Besides the probability ratio for the production of astatine isotopes we calculated the cross section for the production of α particles with energies > 20 Mev through the disintegration of bismuth, using the equation

$$\sigma_\alpha = \frac{\sigma(At^{211})}{\Sigma W_n K_n}.$$

Table IV gives the results obtained for different values of our parameters. The experimental ratio of astatine yields (0.32 to 0.40) is satisfied

TABLE IV

τ, Mev	V, Mev	$\frac{\sigma(At^{210})}{\sigma(At^{211})}$	σ_α ($E_\alpha > 20$ Mev), cm^2
4	13	0.26	$7.3 \cdot 10^{-25}$
4	16	0.27	$6.3 \cdot 10^{-25}$
4	19	0.33	$5.0 \cdot 10^{-25}$
4.5	13	0.34	$6.3 \cdot 10^{-25}$
5	10	0.37	$5.7 \cdot 10^{-25}$
5	13	0.41	$5.4 \cdot 10^{-25}$
5	16	0.43	$5.0 \cdot 10^{-25}$

by values of τ from 4.5 to 5 Mev and low V (10 to 13 Mev). This is higher then the value of 4 Mev obtained by Perfilov and Ostroumov[11] from the averaged spectrum of α particles emitted by Bi and W when bombarded by 460-Mev protons. Figure 2 compares our calculated curves with

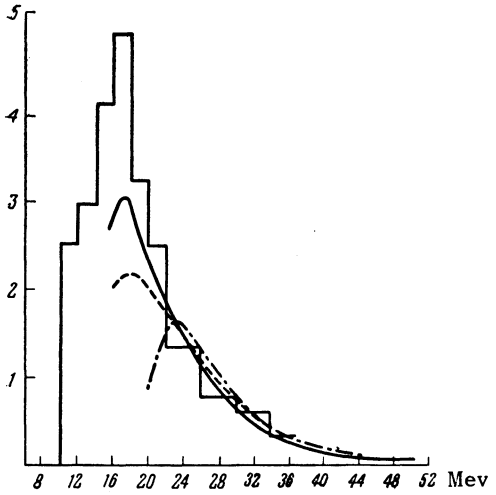

FIG. 2. Spectrum of fast α particles computed according to the yields of At^{210} and At^{211} from bismuth. Solid curve: $\tau = 5$ Mev, V = 13 Mev. Dashed curve: $\tau = 4.5$ Mev, V = 13 Mev. Dots and dashes: $\tau = 4$ Mev, V = 19 Mev.

their histogram. For the purpose of the comparison the curves are normalized to an identical area for the sum of α particles with $E_\alpha > 20$ Mev. The histogram (shifted 2 Mev to the right in accordance with a correction by the authors) fits between the curves for $\tau = 4.5$ and 5.0 Mev with V = 13 Mev. Figure 2 also gives the curve for

τ = 4 Mev and V = 19 Mev, which also satisfies the yield ratio of astatine isotopes. The difference between the peak of this curve and the corresponding experimental point does not at present justify rejection of the curve if we take into account the inadequate statistical information in connection with experiments using photographic emulsions for this range of α-particle energies, and the new results[12] which indicate the existence of a second maximum in the α spectrum under similar conditions.

Although the shape of the α spectrum according to the astatine yield agrees with other data, there is considerable disagreement with experiment in the calculation of the quantity of α particles. Perfilov and Ostroumov[11] obtained 1.5 to 1.6×10^{-25} cm^6 for the cross section of α-particle production with $E_\alpha > 20$ Mev, whereas our computations from the yield of At^{211} give a result which is 3 to 4 times larger (Table IV). Our result is not very sensitive to the assumed spectral shape and we cannot expect it to be reduced for more precise parameters. We can attempt to attribute the discrepancy to inaccurate calculation and the superimposing of experimental errors in the three independent investigations which are used for comparison. But this is evidently not the only possible explanation and the question requires further investigation.

Our results show that the yield ratio of the two heavy astatine isotopes does not depend essentially on the proton energy. This result (like the low value of the barrier V) apparently indicates that α particles with E > 20 Mev appear at a stage of nuclear excitation which is far from a quasi-equilibrium state. A purely evaporational mechanism for the production of the α particles would be an oversimplification. Specifically, we cannot exclude from the astatine-producing process the participation of α particles formed in a cascade process. The latter may be one of the causes of the high value of τ.

THE PRODUCTION OF ASTATINE FROM LEAD

Very pure lead was used. Spectral analysis indicated the presence of 0.01% of bismuth. By chemical analysis, the uranium content was estimated to be < 0.005% and the thorium content < 0.001%. In control experiments we used lead which we had prepared out of "Kalbaum" lead acetate. This material was converted into lead chloride and was recrystallized; the lead was then extracted by electrolysis of a hot aqueous solution, using very pure graphite electrodes. After being washed, the lead was melted in an atmosphere of electrolytic hydrogen. Spectral analysis of the purified lead revealed ~0.001% of bismuth. Chemical analysis did not reveal the presence of uranium or thorium within the limits of experimental sensitivity (U content < 10^{-3}%; Th content < 5×10^{-4}%).

The procedures for bombardment and separation of the astatine were the same as those used in the experiments with bismuth, except for the preliminary operation of precipitating lead with hydrochloric acid and longer bombardment of the sample (from 1 to 2 hrs). In each experiment we obtain α activity with a half-life of 7 to 8 hrs, which corresponds to At^{211}. Shorter periods were at the limit of detection. The astatine activity per gram of lead was considerably smaller than for bismuth; in the best case we obtained 1.9×10^4 decays/min when converted to continuous bombardment. We did not observe any dependence of the astatine yield on the purity of the original material.

The observed effect could not be caused by bismuth impurity, which would have had to be ~0.4% and thus much larger than the actual impurity. Astatine production due to uranium impurity is also excluded; this would have had to be 0.7%. An estimate of the possible influence of thorium impurity is a more complicated matter. The computed effect of thorium impurity comprises not more than one fourth of the observed activity of astatine from lead. However, we are inclined to attribute the astatine activity to reactions in lead nuclei; the actual amount of thorium impurity is unknown and may be smaller than the figure mentioned and, on the other hand, the observed At^{211} activity is a lower limit because of possible loss in chemical separation.

The formation of At^{211} from the heaviest lead isotope Pb^{208} is possible only through the capture of light nuclei resulting from the disintegration of lead. Considering lithium nuclei in the first place, the reaction can be written as Pb^{208}(Li, kn)At^{211}, although the nuclei of beryllium and other heavier elements may also participate. The cross section of At^{211} production for 480-Mev protons is estimated at 0.4 to 0.8×10^{-31} cm^2.

SEARCH FOR OTHER SECONDARY REACTIONS

For the purpose of detecting the capture of carbon nuclei we performed exploratory experiments to study the production of astatine from gold bombarded by 480-Mev protons, thus increasing the charge by 6 units. A small amount of α activity was detected (about 10 pulses/min against a back-

ground of 2 pulses/min) decaying with a half-life of 7 to 8 hrs. This small effect, which represents a cross section $\sim 10^{-33}$ cm^2, did not enable us to determine the source of the activity, as thorium impurity amounting to 5×10^{-6}% could result in the same activity. Despite special purification of the gold, thorium impurity to this extent cannot be excluded. Thus the sought reaction of carbon capture (in our case C^{14} and heavier isotopes) lies at the limit of observation and cannot be established with certainty at the bombarding energies which were used.

A similar result was obtained in our investigation of the production of iodine from silver (carbon capture) bombarded by 480-Mev protons. In this instance also the small iodine yield (16 pulses/min, representing $\sigma \approx 5 \times 10^{-33}$) did not permit us to establish its production unambiguously as the result of the sought reaction.

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ENERGY SPECTRUM AND ANGULAR DISTRIBUTION OF π^+ MESONS, PRODUCED IN PROTON-PROTON COLLISIONS AT 660-670 Mev

A. G. MESHKOVSKII, Ia. Ia. SHALAMOV, and V. A. SHEBANOV

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The energy spectrum of π^+ mesons produced in p – p collisions by 670 Mev protons were measured for observation angles 19°30′, 38°, and 56°. It was found that, in the c.m.s., the shape of the π^+-meson spectrum for the $pp \rightarrow pn\pi^+$ reaction depends on the emission angle. The angular distribution of π^+ mesons produced in 660-Mev p – p collisions is given, in the c.m.s., by Eq. (1). The value found for the total cross-section is $(14.4 \pm 1.2) \times 10^{-27}$ cm^2.

1. INTRODUCTION

THE energy spectrum and the absolute yield of π^+ mesons, produced in p – p collisions at 660 Mev, were first measured by Sidorov[1] in emulsions, at five values of the observation angle from 60° to 120°. Meshcheriakov et al.[2] studied the relative spectrum of π^+ mesons at 24° by means of magnetic analysis. Meshkovskii et al.[3] measured the absolute yield of π^+ mesons at 29°, 46°, and 65°