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Formation of Radium E and Polonium by Deuteron Bombardment of Bismuth

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The direct production of RaE and Po by deuteron bombardment of bismuth has been reinvestigated. Present results indicate that as the deuteron energy is lowered from 8.8 to 7.3 Mev, the production ratio of RaE/Po increases 25 percent but as the deuteron energy is dropped from 7.3 Mev to 5.5 Mev, the ratio more than doubles. The trend is in agreement with the Oppenheimer-Phillips reaction in which RaE is produced by a *d*, *p* reaction and Po is produced by a *d*, *n* reaction. Assuming these reaction cross sections to be respectively proportional to Bethe's OP penetration factor and to the Gamow-Condon-Guerney penetration factor, best agreement of the variation with energy of the ratios of these cross sections is obtained with a nuclear radius of $2.0A^{1/3}\times10^{-13}$ cm for bismuth.

INTRODUCTION

THE disintegration products of bismuth under bombardment by deuterons with energy in the region of 5 to 9 million electron volts has been studied experimentally and two different results were found.[1,2] Since bismuth with its single isotope and high atomic number affords an excellent opportunity for the comparison of two competing processes under deuteron bombardment—proton emission and neutron emission—the problem was reinvestigated experimentally at the University of Michigan Physics Department in 1940. Because of the war, the results have only just now been interpreted and are herewith reported.

THE NUCLEAR REACTIONS

The nuclear reactions have been previously reported and there is a *d*, *p* and a *d*, *n* reaction which may be described by means of three processes:

I. ${}_1d^2+{}_{83}\mathrm{Bi}^{209}\rightarrow{}_{83}\mathrm{RaE}^{210}+{}_1p^1$,

II. ${}_1d^2+{}_{83}\mathrm{Bi}^{209}\rightarrow{}_{84}{}^*\mathrm{Po}^{211}\rightarrow{}_{83}\mathrm{RaE}^{210}+{}_1p^1$,

III. ${}_1d^2+{}_{83}\mathrm{Bi}^{209}\rightarrow{}_{84}{}^*\mathrm{Po}^{211}\rightarrow{}_{84}\mathrm{Po}^{210}+{}_0n^1$.

The first two reactions have the same end products and so may not be distinguished by measurement of only the end products. Process I is an Oppenheimer-Phillips reaction[3] in which the deuteron splits up outside the nucleus with the neutron being absorbed in the final nucleus which is identical with the compound nucleus. Formally, reaction II appears the same as I but here an intermediate compound nucleus is formed and the proton emission must compete with the more probable neutron emission as in III. If III is more probable than II, only I and III will occur and these two are distinguishable by a fortunate set of circumstances. The RaE formed in I decays by β-ray emission with a half-life of 5.0 days into polonium and the Po thus formed, as well as the Po produced in III, decays with a lifetime of 136 days by the emission of α-particles. Thus, by the measuring of the α-particle activity as it changes, the amounts of each product may be determined. The number of Po nucleii N_2 present at any time is given by

$$N_2=[\lambda_1N_1^0/(\lambda_1-\lambda_2)][\exp(-\lambda_2t)-\exp(-\lambda_1t)]+N_2^0\exp(-\lambda_2t). \quad (1)$$

N_1^0 is the number of RaE nucleii formed by the bombardment in reaction I (and II) while N_2^0 is the amount of Po nucleii present at $t=0$ (at the instant of bombardment) in reaction III. λ_1 and λ_2 are, respectively, the decay constant of RaE

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[1] Hurst, Lantham, and Lewis, Proc. Roy. Soc. **174**, 126 (1940).

[2] Cork, Halpern, and Tatel, Phys. Rev. **57**, 348 (1940); **57**, 371 (1940).

[3] H. A. Bethe, Phys. Rev. **53**, 39 (1938).

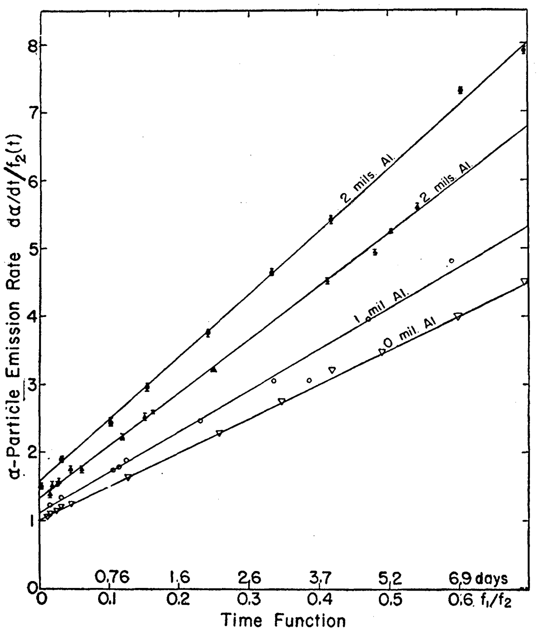

FIG. 1. Straight-line plot of alpha-particle emission as a function of time for the various targets with deuteron bombarding energy. Ordinates should be multiplied by 1.10×10^8, 1.60×10^8, 10.0×10^8, and 3.06×10^8 for the 0, 1, 2, 2 mil curves, respectively.

(1.60×10^{-6} second^{-1}) and Po (5.89×10^{-8} second^{-1}). N_1^0 and N_2^0 may be found from the experiment by integration or by differentiation of (1). For the latter method, the alpha-particle emission rate at any time is given by $d\alpha/dt = -\lambda_2 N_2$ which can be combined with (1) to give

$$\frac{d\alpha}{dt}\Big/ f_2(t) = N_1^0[f_1(t)/f_2(t)]+N_2^0; \qquad (2)$$

$$f_1(t) = [\lambda_1\lambda_2/(\lambda_1-\lambda_2)] \times[\exp(-\lambda_2 t)-\exp(-\lambda_1 t)],$$

$$f_2(t) = \lambda_2 \exp(-\lambda_2 t).$$

Expression (2) is the equation of a straight line and so if the α-particle rate divided by f_2 is plotted as ordinate against f_1/f_2 as abscissa, the intercept N_2^0 gives the yield in III and the slope N_1^0 is a measure of the yield in I (and II). Figures 1 and 2 show plots of the data in this form. The vertical lines through each point are a measure of the statistical fluctuation of the number of counts. This fluctuation is taken as $N^{\frac{1}{2}}$ where N is the total number of alpha-particles measured in a given interval.

THE OBJECT OF THE EXPERIMENT AND GENERAL PROCEDURE

For a given target of bismuth, it is possible to compare the cross sections for deuteron bombardment of the two types of reactions, emission of a proton and emission of a neutron. Generally, when a compound nucleus is formed at excitations below the barrier height, it is considerably more probable that a neutron is emitted than a proton because, for one thing, the neutron does not have to penetrate the potential barrier. However, if the mechanism proposed by Oppenheimer and Phillips prevails, it is possible for the d, p reaction to be more probable. A test of whether or not the OP process occurs is to compare its cross section with an ordinary reaction as the deuteron bombarding energy is varied, the OP process diminishing relatively as the top of the deuteron barrier is approached. In this experiment, the energy of the deuterons was varied by interposing aluminum foils between

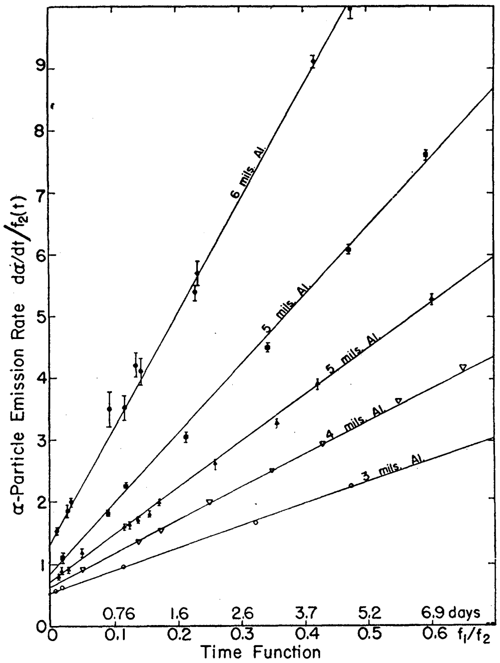

FIG. 2. Straight-line plot of alpha-particle emission as a function of time for the various targets with lower bombarding energy. The ordinates should be multiplied by 2.00×10^8, 2.00×10^8, 10.0×10^8, 10.0×10^8, 263×10^8 for the 3, 4, 5, and 6 mil curves, respectively. In comparing Fig. 2 to Fig. 1, it is seen that there is greater slope divergence in Fig. 2 although the intercept spread is over the same range. This increase in slope indicates relatively more N_1(RaE) with lower energy.

TABLE I. Experimental results.

Mils of Al	Deuteron energy Mev	N_1^0 $\times 10^{-7}$	N_2^0 $\times 10^{-7}$	Numbers of deuterons in coulombs	Bombardment time-hr.	$N = N_1^0/N_2^0$	R	$\sigma_{dp} \times 10^{28}$ cm²	$\sigma_{dn} \times 10^{28}$ cm²	R_{HLL}	R_{CHT}
0	8.7	55.2	11.3	0.012	0.58	4.9	5.0	34	6.8	4.3	4.9
1	8.2	36.9	7.3	0.015_6	0.5	5.1	5.2	18	3.4	5.0	5.0
2	7.8	20.9	3.7	0.016_8	1.0	5.6	5.7	$9._2$	1.6	4.9	6.0
2	7.8	29.9	5.3	0.015_0	0.5	5.6	5.7	15	2.6		
2*	7.8	7.8	1.3_1	0.027_7	1.5	5.8	6.0	$6._3$	1.0_5	6.3	
3	7.3	17.8	2.7_8	0.016_6	0.83	6.2	6.3	8	1.3_0		8
4	6.8	27.2	3.2	0.040_4(?)	1.8	8.1	9.5	5	0.5		17
5	6.3	7.5	0.72	0.036_7	2.0	$10._2$	$10._9$	1.5	0.14	6.5	
6	6.3	11.3	0.83	0.056_5	2.0	13	14	1.4	0.10		
6	5.8	0.52	0.033	0.07_2	4.0	$13._6$	16	0.5_4	0.03_4	—	

* Target of approximately 3.2 mg/cm² of Bi on Al.

the beam and the bismuth target. Hence, for a given energy, a bismuth target was bombarded and the rate of emission of alpha-particles was measured from the time the bombardment stopped and plotted as already explained. Then N_1^0 and N_2^0 which are the end products of the *d*, *p* and *d*, *n* reaction, respectively, may be found and so their ratio for each energy. Since for each target the deuteron energy and distribution is identical, an excellent value of the ratio of these two quantities $N = N_1^0/N_2^0$ may be found. This ratio must be corrected for the time of bombardment especially for the weak samples which have both a long bombardment time and a large ratio. The reason is that the radium E is always decaying into polonium and so at the end of a bombardment there will, in general, be less radium E and more polonium thereby giving a smaller ratio. The true ratio of cross sections is to a first and sufficient approximation:

$$R = \frac{N}{1 - \frac{1}{2}\lambda_1 T(N+1)} \tag{3}$$

where λ_1 is the decay constant of radium E and T is the bombardment time. This correction amounts to 1 percent for an N of 5.6 and a bombardment of 0.5 hour; and to 9 percent for a bombardment of 2 hours with an N of 13. These values with N and R and T are shown in Table I.

There are two types of targets which have been used in this experiment and both are "thin." HLL[1] used evaporated layers of bismuth on thin aluminum foils with a total thickness of about 1 air-cm. The second procedure[2]—used here—employs a physically thick piece of bismuth for the target but makes use of the short range 3.8 air-cm polonium alpha-particles compared to the long range deuterons to secure an essentially thin target. Since the alpha-particles had to travel one centimeter before entering the ionization chamber grid to be registered, the only ones counted must originate within 2.8 air-cm of the surface of the bismuth target. At the deuteron energy limits of 6 and 9 Mev used in this experiment, the deuterons lose, respectively, 0.07 and 0.05 Mev/cm or 0.20 and 0.14 Mev in the effective thickness (2.8 air-cm) of the target. Hence no correction need be made here for the target thickness. There is a disadvantage in using this thick target scheme, for the alpha-particles coming into the chamber have residual ranges extending from zero to 2.8 air-cm and so fluctuations in over-all counter sensitivity give variations in counting rates. However, a change in counter sensitivity of 1 percent gave a change of less than 1 percent in the counting rate. Before each count, the amplifier sensitivity was set to within 1 percent of a fixed value and generally the statistical fluctuations in counts was about 1 percent so that this error does not affect the readings more than the statistical fluctuations of the counting. The advantage in this type of thick target is that the target is simple to prepare and there is little danger in burning it out during bombardment or breaking it during the subsequent measurements.

Each bombardment was monitored with a current integrator to obtain absolute cross section. However, since the cyclotron was never considered to give an identical bombardment for different runs, the absolute cross section is not

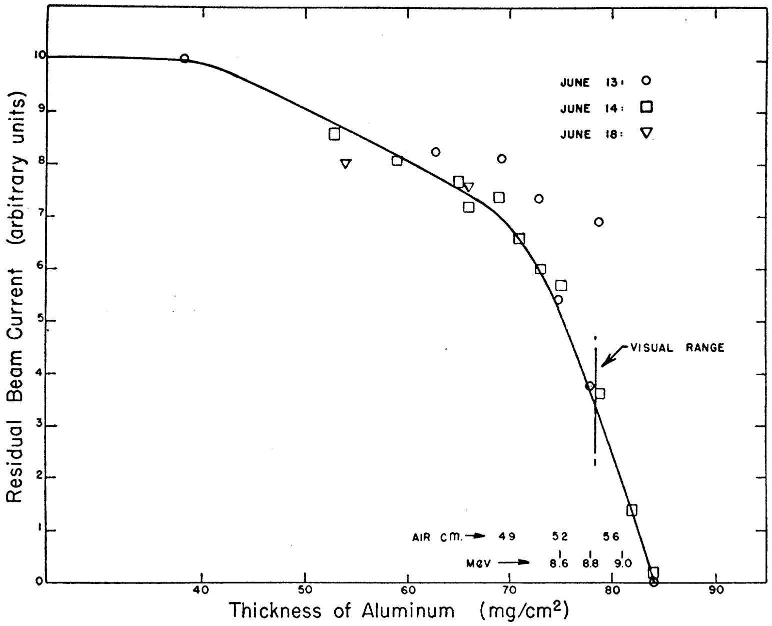

FIG. 3. Plot of residual current of deuteron beam of cyclotron through different thicknesses of aluminum foil.

considered to be a reliable result although the trend should be fairly consistent.

The energy of the deuteron beam was estimated visually by measuring the range in air of the beam, the mean range being taken as the point along the beam at which the brilliancy of an irradiated fluorescent screen started to diminish rapidly. This measurement was made as a check on a residual range determination in which the residual beam current to a Faraday cup was measured as the beam passed through aluminum foils of various thickness, Fig. 3. The air equivalent of aluminum was taken as 1.52 milligrams per cm^2 per air-cm and corrected according to Livingston and Bethe. The effective maximum deuteron range is taken as 54 air-cm or 8.8 Mev. Although, the beam is not monoenergetic, the trend of the results should not be seriously affected as the cross section is changing so rapidly that for any experiment only the high energy tail is efficacious in making the transmutation. The straggling in energy for a 5- and 9-Mev deuteron beam is about 0.1 and 0.16 Mev, respectively, while the approximate straggling of the beam in the cyclotron was 0.5 Mev.

RESULTS

Bismuth targets were bombarded with deuterons from zero to 0.006 inch of aluminum in steps of 0.001 inch (6.6 mg/cm^2). These steps decreased the mean energy of the beam by approximately 0.5 Mev per step from a maximum of 8.8 Mev. During bombardment, the beam current was kept to within 70 percent of a norm and the total quantity of deuterons and the time of bombardment recorded. After bombardment, the targets were mounted in front of a parallel plate ionization chamber and linear pulse amplifier arrangement. Counting rates of the emitted alpha-particles were measured at intervals for from five to ten days. All targets showed an initial rate, an increase, and then (if followed long enough) a decrease. The graphs in Figs. 1 and 2 show that each target accurately follows the assumed pattern of formation of RaE and Po. The quantities of these end products, their ratios, and the ratios corrected for finite bombardment time are all shown in Table I. The absolute cross section is roughly computed assuming that all the Bi nucleii in 2.8 air cm (10.6 mg/cm^2) are effective as targets and that the effective solid angle is defined by the circular 1.5-cm opening of the ionization chamber 1 cm from the target under observation. For comparison, the ratios from the two previous measurements are also tabulated R_{HLL}[1] and R_{CHT}[2]. These results are plotted as follows: cross section for formation of Po (σ_{dn}) in Fig. 4 and the corrected ratio R for finite bombardment time, of RaE/Po or σ_{dp}/σ_{dn} in Fig. 5.

SOURCES OF ERROR

A major source of uncertainty in the measurements is the deuteron energy. There are two difficulties. First, the beam had a considerable spread as shown in Fig. 3 and second, the energy varied from time to time. This may be the reason for the disagreement of the two measurements of R with 5 mils of Al. Because so many measurements were taken on each sample, it is thought that the measurement of R on any particular sample has an over-all accuracy of 10 percent at 6 mils to 3 percent at zero mils of Al absorber in the beam.

DISCUSSION

As has been pointed out, the d, p reaction may formally be described by either an OP process, I, or by the formation of a compound nucleus with subsequent proton mission, II. The reaction III is clearly of the latter type. In order to compare the OP process, I, with an "ordinary" process as III, it would be necessary for reaction II to be extremely improbable. However, there are no experimental data by which reaction II may be ruled out compared to reaction III.

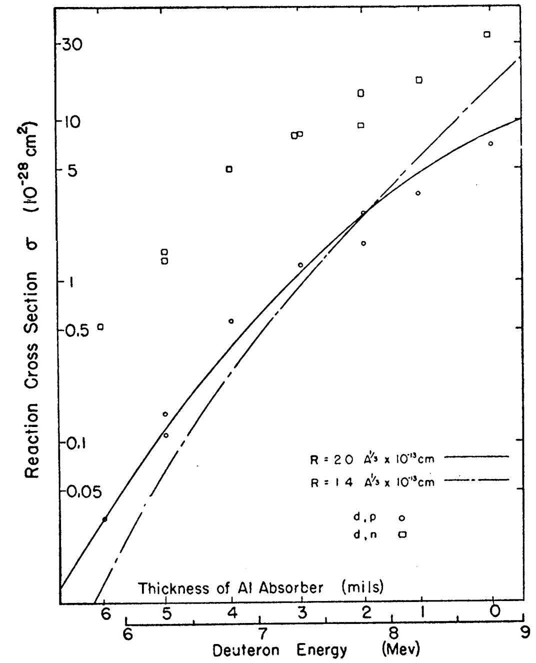

FIG. 4. Plot of cross section of d, n reaction as a function of the energy. The two curves are plotted for the Gamow-Condon-Guerney penetration function for small and large nuclear radius. Both curves arbitrarily fit at about 8 Mev.

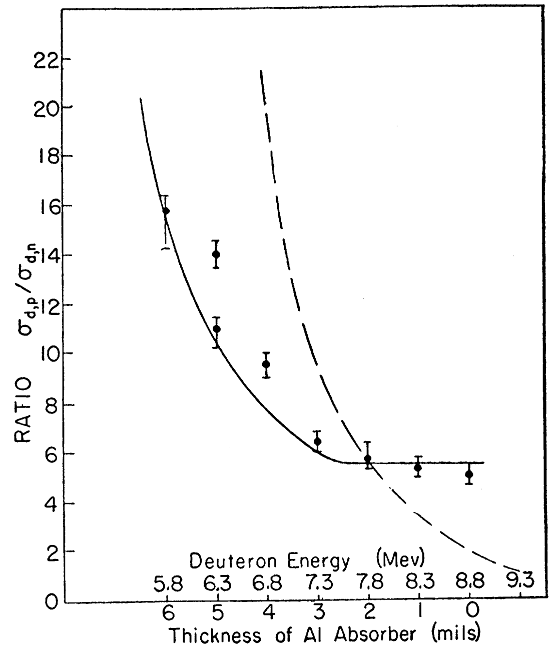

FIG. 5. Plot of ratio of cross sections of d, p and d, n reaction as a function of the deuteron bombarding energy. The curves are the ratio of Bethe's Oppenheimer-Phillips penetration factor to the Gamow-Condon-Guerney penetration factor. The best fit is with the larger nuclear radius of $2.0A^{1/3}\times 10^{-13}$ cm.

It is usually considered that when a heavy compound nucleus is formed, the competition of the various processes is such that the emission of a neutron is considerably more probable than emission of a proton and so reaction III would be more probable than reaction II. This is deduced by Bethe,[4] paragraph 79, in the following manner: For the ordinary type of reaction in which a compound nucleus is formed by deuteron bombardment with subsequent emission of neutrons and protons:

$$\sigma_{dn}=\pi\lambda\!\!\!^{-}{}^2\xi_d P_d l_c{}^2\Gamma_n/\Gamma$$

and

$$\sigma_{dp}=\pi\lambda\!\!\!^{-}{}^2\xi_d P_d l_c{}^2\Gamma_p/\Gamma \qquad (4)$$

where ξ_d is the deuteron "sticking function," P_d is the deuteron penetrability, l_c is the "effective orbital momentum," and Γ is the total width of

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[4] H. A. Bethe, Rev. Mod. Phys. 9, 69 (1937).

all levels of the compound nucleus in the energy region in question. All these quantities are functions of the incoming particles leaving Γ_n, Γ_p, the partial widths of the states with emitted particles n or p in the final residual nucleus. Hence,

$$\sigma_{dn}/\sigma_{dp}=\Gamma_n/\Gamma_p. \tag{3}$$

These partial widths depend on the total number of available states and on the penetrabilities of the emitted particles both of which in turn depend upon available energy. The difference in this available energy is

$$U_n-U_p=\epsilon_- - 0.8\text{ Mev}+B_p,$$

which is expression 40 from Bethe,[3] where ϵ_- is the energy of the β-ray of RaE (in this particular case 2 Mev), the 0.8 Mev is the mass difference of neutron and hydrogen atom, and B_p is the proton barrier height (5 Mev) whence

$$U_n-U_p\sim 6\text{ Mev}.$$

Hence, there is considerably more energy available for the neutron emission than for the proton emission and since the partial widths depend upon the energy available, it is seen that Γ_n should be greater than Γ_p and, therefore, from (3) the cross section for emission of the neutron should be greater than the cross section for the emission of the proton. While there is not enough knowledge on the details of these nucleii available, a probable estimate of this ratio may be obtained from statistical considerations. Bethe (41)[3] shows that the ratio of the neutron and proton partial widths is given by

$$\Gamma_n/\Gamma_p=\exp(U_n-U_p)/T.$$

The nuclear temperature T for bismuth for this type of excitation will be approximately 1.3 Mev and so the ratio of the cross sections would be a rather large number e^4 which would indicate that reaction III is considerably more probable than reaction II and thus the experiment is measuring only reaction I and III. Assuming then that this is true, it should be noted that Bethe's critiques[3] for observing the OP process are fulfilled in this reaction, i.e., the deuteron bombarding energy is below the barrier height minus the deuteron binding energy (2.2 Mev). The barrier height of bismuth for deuterons is 9.8 and 14.3 Mev for the large and small nuclear radius, respectively.

The only factor used here in the theoretical treatment of the comparison of the Oppenheimer-Phillips type of reaction to the ordinary type of reaction are the penetrability factor. These are also the most rapidly varying factors with respect to energy and it is usually considered that the ratio of the cross sections (γ) is given by the ratio of the penetrabilities. Hence

$$\gamma=\sigma_{\text{OP}}/\sigma_{\text{GCG}}\sim P_{\text{OP}}/P_{\text{GCG}},$$

where the subscript OP refers to the Oppenheimer-Phillips penetrability function and GCG refers to the Gamow-Condon-Guerney type of penetrability for the deuteron.

In fitting the theoretical γ to the experiment, the most important parameter is the nuclear radius. Near the barrier height, this is especially important as the theory predicts that $\gamma=1$ at the barrier height minus the binding energy of the deuteron. For a radius of $2.0A^{\frac{1}{3}}\times 10^{-13}$ cm and $1.4A^{\frac{1}{3}}\times 10^{-13}$ cm, this occurs for $Z=83$ at 7.6 and 12.1 Mev, respectively. In Fig. 5, the data have a more slowly varying region in the neighborhood of 7.6 Mev which is what the theory predicts for the larger nuclear radius. The γ's for these two values are plotted also in Fig. 5. The values for the smaller radius are taken from Volkoff's[5] discussion of Bethe's OP theory. The theoretical curves are fitted to the experimental values at 7.6 Mev. The trend of the results are in general agreement with an OP process of finite nuclear radius and the radius best matching these results is the larger $2.0A^{\frac{1}{3}}\times 10^{-13}$ cm. Of further interest, it is seen that d, n follows rather closely the P_{GCG} function and so the effect of the other factors which enter in the cross sections other than the penetration factors varies slowly compared to them.

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[5] G. M. Volkoff, Phys. Rev. **57**, 866 (1940).