(p, pn) and $(p, 2p)$ Reactions of Ce¹⁴² at Proton Energies from 0.4 to 3.0 Bev*

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Cross sections are reported for (p, pn) and $(p, 2p)$ reactions of Ce¹⁴² with protons of 0.4, 1.0, 2.2, and 3.0 Bev. To evaluate the contribution of proton and neutron evaporation from excited Ce¹⁴² nuclei to the observed cross sections, this evaporation competition was studied by means of cross-section measurements on the reactions Ba¹³⁸(α ,n) and Ba¹³⁸(α ,p) with α particles of 19-to 40-Mev kinetic energy. The ratio σ_{α} , α/σ_{α} , p is between 2.4 and 4.4 in this region as compared with a ratio of 1.58 \pm 0.14 for $\sigma_{p,pn}/\sigma_{p,2p}$ at 0.4 Bev. It is concluded that at this energy the (p, pn) and $(p, 2p)$ reactions proceed at least partially by a pure knock-on mechanism. This conclusion is supported by comparison of the data with recent Monte Carlo calculations of intranuclear cascades. With increasing proton energy both $\sigma_{p, 2p}$ and $\sigma_{p, pn}$ are found to decrease, the former more rapidly than the latter. This energy dependence is discussed in terms of the elementary nucleonnucleon cross sections. The Monte Carlo calculations mentioned agree within their rather poor statistical accuracy with the measured cross-section ratios and with the energy dependence of the cross sections, but predict cross-section values about a factor of three too small.

INTRODUCTION

" 'N studies of the interactions of complex nuclei with Γ high-energy protons, it has been observed^{1,2} that rather simple reactions occur with relatively high cross sections and that these cross sections are not strongly energy dependent in the region from a few hundred to a few thousand Mev. Among these simple reactions the so-called (p, pn) and $(p, 2p)$ reactions, i.e., interaction in which \overline{A} decreases by one unit and \overline{Z} either remains the same or decreases by one, are of particular interest, because in the energy range mentioned they account for several percent of the geometric cross sections of the target nuclei. $3-8$

To explain the large cross sections observed for (p, pn) reactions, it has been suggested³ that they proceed by a pure knock-on mechanism. By this is meant a collision of the incident proton with a nucleon in the nucleus, followed by the escape from the nucleus of both collision partners without further interactions. An additional restriction is that the excitation energy left by the creation of the nucleon hole must be less than the binding energy of the most loosely bound neutron.

In a second possible mechanism for a (p, pn) reaction, one nucleon only is emitted during the knock-on cascade, and the other is subsequently evaporated.

The simplest (and most probable) process of this type involves a single glancing collision of the incident proton with a nucleon, accompanied by the transfer of the order of 10—²⁰ Mev to the nucleon and followed by the escape of the proton. Subsequent evaporation of a neutron then completes the (p, pn) reaction whereas evaporation of a proton would lead to a $(p, 2p)$ reaction.

The present study was designed to lead to a decision between the two mechanisms outlined, at least in the region of 400 Mev. At higher energies it was hoped that it would throw some light on the effects of mesonic processes. Leaving the latter aside for the moment, it seemed clear that the two mechanisms should lead to different values for the ratio of (p, pn) to $(p, 2p)$ cross sections for a given target nuclide (Z^A) . For the second mechanism, which goes via a compound nucleus $(Z^A)^*$ of relatively low excitation (10—20 Mev), the crosssection ratio would be determined by the relative probabilities of neutron and proton evaporation from such a compound nucleus. For a pure knock-on mechanism, on the other hand, the cross-section ratio would be in first approximation the n/p ratio in the nucleus, modified by the ratio of $p\n- n$ to $p\n- p$ collision cross sections.

The target nucleus chosen for this investigation was Ce¹⁴². The product of the (p, pn) reaction is the 32-day β^- emitter Ce¹⁴¹; that of the $(\rho,2\rho)$ reaction is 3.7-hour. La¹⁴¹ which decays by β ⁻ emission to Ce¹⁴¹. The competition between neutron and proton evaporation from a $Ce¹⁴²$ compound nucleus was studied by measurement of the cross sections of the reactions $Ba^{188}(\alpha,n)Ce^{141}$ and $Ba^{138}(\alpha, p)La^{141}$ with helium ions of energies up to about 40 Mev.

EXPERIMENTAL

Proton Bombardments

Cerium in the form of $CeO₂$ was bombarded in the circulating beam of the Cosmotron at proton energies of 1.0 Bev and above. Bombardment techniques at the

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¹ D. H. Templeton, *Ammual Review of Nuclear Science* (Annual Reviews, Inc., Stanford, 1953), Vol. 2, pp. 93–104.
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^{94, 727 (1954).&}lt;br>³ E. Belmont and J. M. Miller, Phys. Rev. 95, 1554 (1954).

⁴ G. D. Wagner and E. O. Wiig, Phys. Rev. 96, 1100 (1954). ⁵ J. B. Cumming, Ph.D. thesis, Columbia University, 1954

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A. A. Caretto, Jr., and E. O. Wiig, Phys. Rev. 103, 236 (1956). R. W. Fink and E. O. Wiig, Phys, Rev. 94, 1357 (1954).

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University, 1957 (unpublished).

3rookhaven Cosmotron have previously been described.^{2,9} The 400-Mev data were obtained by use of the Nevis cyclotron. All targets were prepared by forming a slurry of spectroscopically pure $\text{CeO}_2{}^{10}$ in acetone and allowing this mixture to settle in a large glass chimney onto a piece of 0.003-inch aluminum foil. The excess acetone was siphoned off and a small quantity of a dilute solution of Duco cement in acetone was placed on top of the $CeO₂$ mat. This was allowed to evaporate to dryness. The resulting mat of $CeO₂$ on aluminum was quite durable and could be cut to the desired shape. The thickness was about 20 mg/cm', uniform to about $\pm 20\%$ over an area of about 25 sq in. and better than this over smaller areas. The effective beam intensities were in the range of 10^{12} to 10^{13} protons per minute incident on the target.

The target consisted of an aluminum- $CeO₂$ mat plus an additional 3-mil aluminum foil to be used to monitor the proton beam by measurement of the yield of Na²⁴ from the known excitation function of the $Al^{27}(p,3pn)Na^{24}$ reaction.¹¹ The two foils were cut to the same area and the leading edges were accurately aligned. In one run at 400 Mev, a different monitoring technique was used. A uniform mixture of the oxides of cerium and aluminum, weighing about 10 mg/cm², was irradiated and the $Na²⁴$ produced from the aluminum was chemically separated from the target solution and used to monitor the proton beam.

After irradiation the aluminum monitor foil was separated from the rest of the target and reserved for the Na²⁴ determination. The aluminum-CeO₂ mat was dissolved in concentrated HCl with the addition of a few crystals of KI to aid the dissolution of $CeO₂$; the I_2 produced was reduced with 30% H_2O_2 . Ten milligrams of inert lanthanum carrier were added, the solution was made up to a known volume, and a known aliquot was taken and reserved for the cerium determination by standard analytical methods. The cerium and lanthanum were separated from the bulk of the other activities by a fluoride precipitation. The fluoride precipitate was dissolved in boric and nitric acids, and cerium and lanthanum were reprecipitated as hydroxides. Solution of the hydroxides in $8N$ HNO₃ was followed by oxidation of cerium with $BrO₃^-$, and separation of the cerium from the lanthanum was achieved by the solvent extraction of the cerium in the plus four state into methyl isobutyl ketone according to the procedure outlined by Glendenin et al.¹² The 3.7-hour La¹⁴¹ in the lanthanum fraction was allowed to decay completely to the 32-day Ce¹⁴¹ daughter. This latter activity was then chemically isolated from the lanthanum fraction and the Ce¹⁴¹

counted. The cerium activities produced from the reactions on Ce^{142} and the Ce^{141} produced by the decay of La'4' were both counted with end-window proportional counters.

All samples were mounted as $Ce_2(C_2O_4)_3.9H_2O$ on filter paper, mounted on aluminum cards, covered with rubber hydrochloride $(1-2 \text{ mg/cm}^2)$ and counted for times sufficiently long to resolve the 32 -day Ce¹⁴¹ from 140 -day Ce^{139} . Since all the activity measurements were done on Ce¹⁴¹ samples, the determination of the $\sigma_{\text{Ce}^{141}}/\sigma_{\text{La}^{141}}$ ratios was free of counting-efficiency corrections. The cross sections were calculated taking into account the yield of Na'4 from the aluminum monitor, chemical yields of the cerium and lanthanum recovered, the growth and decay of the Ce^{141} and La^{141} during bombardment,¹³ the decay of the La¹⁴¹ before chemical separation, and the usual saturation and countingefficiency corrections, the latter including self-absorption and self-scattering corrections experimentally determined for Ce¹⁴¹ radiations in ceric oxalate precipitates.

TABLE I. Cross sections for the production of Ce¹⁴¹ and La¹⁴¹ in the bombardment of Ce¹⁴² with protons between 0.4 and 3.0 Bev.⁸

^a The errors given for the individual cross sections are deviations of the last duplicate determinations from the last column represent the estimated 12% uncertainty in the absolute value of the cross-section ratios.

Alpha-Particle Bombardments

Metallic barium was irradiated in the deflected beam of the 60-inch cyclotron. The targets were prepared by the evaporation of metallic barium on 0.003-inch aluminum foil to a thickness of about 0.5 mg/cm². A thin coating of aluminum was evaporated on top of the barium to protect it from oxidation. Four to six of these barium films were irradiated in a stack, with aluminum absorbers suitably interposed to obtain various alpha-particle energies below the 42-Mev maximum energy of the beam.

The alpha beam was monitored by measurement of the At²¹¹ activity produced by the Bi²⁰⁹(α ,2n)At²¹¹ reaction in a bismuth foil incorporated in the stack. The excitation function of this reaction has been measured. '4 In one experiment the alpha beam was also measured more accurately by means of a calibrated ion collector¹⁵; this gave substantially the same result as the bismuth monitor. '

⁹ Wolfgang, Baker, Caretto, Cumming, Friedlander, and Hudis
Phys. Rev. 103, 394 (1956).
¹⁰ "Specpure" brand CeO₂ obtained from Johnson, Matthe

and Company, Ltd., London.
¹¹ Friedlander, Hudis, and Wolfgang, Phys. Rev. 99, 263 (1955).

¹² Glendenin, Steinberg, Flynn, and Buchanan, Anal. Chem. 27, 59 (1955).

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¹⁶ R. H. Schuler and A. O. Allen, Rev. Sci. Instr. 26, 1128 (1955).

The target foils were dissolved in HCl, and after addition of 10 mg of Ce and 10 mg of La carrier, the same chemical separations were performed as in the high-energy proton experiments. An aliquot of the target solution was removed for barium analysis by flame photometry. The measurements and cross section calculations were performed in the manner described for the proton irradiations.

RESULTS

The cross sections for the (p, pn) and $(p, 2p)$ reactions on Ce^{142} are given in Table I and are illustrated graphically in Fig. 1. All the values given are averages of two determinations and the errors given are the devi-

FIG. 1. Cross sections for the formation of Ce¹⁴¹ and La¹⁴¹ by the interaction of Ce^{142} with high-energy protons.

ations from the averages. The cross sections for the (α,n) and (α,β) reactions on Ba¹³⁸ are given in Table II and illustrated in Fig. 2. Here the values given are averages of between two and four determinations (with their standard deviations), except the data at 29 Mev which are based on only one experiment. Errors in these determinations arise from a number of sources. Errors in relative monitoring of proton beams by. use of aluminum foils amount to only a few percent at a given energy. The absolute value of the $Al^{27}(p,3pn)$ cross section was taken as 10.5 mb at the proton cross section was taken as 10.5 mb at the proton
energies used $(0.4$ to 3 Bev),^{16,17} and is uncertain by

FIG. 2. Cross sections for the formation of Ce¹⁴¹ and La¹⁴¹ by (α ,n) and (α ,p) reactions of Ba¹³⁸. The dotted curve represent the sum of the (α, n) and (α, p) cross sections. The dashed curve is the total cross section for capture of α particles by Ba¹³⁸ com-
puted according to reference 20 for $r_0 = 1.5 \times 10^{-13}$ cm.

about $\pm 10\%$. Uncertainties in counting efficiency, backscattering, air and window absorption, self-absorption, and self-scattering were estimated to be about $\pm 15\%$. Errors arising from the resolution of decay curves may be as large as 10% , and the chemical yield determinations were accurate to about 5% . As a result of all these sources of error it is believed that the reported cross sections are accurate to about $\pm 25\%$. However, the ratios $\sigma_{p, pn}/\sigma_{p, 2p}$ and $\sigma_{\alpha, n}/\sigma_{\alpha, p}$ are subject only to the last two sources of error mentioned and should be reliable to about $\pm 12\%$ as indicated in the last columns of Tables I and II.

DISCUSSION

0.4-Bev Data

From the data presented, the most clear-cut deduction about the dominant mechanism of (p, pn) and $(p, 2p)$ reactions can be made at the lowest energy investigated, 0.4 Bev, where meson effects are unimportant. Here a value of about 1.6 was found for the

TABLE II. Cross sections for the production of Ce¹⁴¹ and La¹⁴¹ by the bombardment of Ba¹³⁸ with helium ions.⁸

Kinetic energy (Mev)	σ Ce ¹⁴¹ (mb)	σ La ¹⁴¹ (m _b)	σ Ce ¹⁴¹ / σ La ¹⁴¹
19.3 25.5 29.0 36.0 41.0	$95+6$ 798 685 $427 + 3$ $296 + 25$	$40.3 + 0.3$ $279 + 27$ 223 124 $+$ 1 67.6 ± 3.9	$2.4 + 0.3$ $2.9 + 0.3$ $3.0 + 0.4$ $3.4 + 0.4$ $4.4 + 0.5$

^a The errors given for the cross sections are standard deviations of the last column represent the last column represent the last column represent the estimated 12% uncertainty in the absolute values of the cross-section ratios.

¹⁶ R. L. Wolfgang and G. Friedlander, Phys. Rev. 96, 190

^{(1954);} 98, 1871 (1955). ' Cumming, Swartz, and Friedlander, Bull. Am, Phys. Soc. Ser. II, 1, 225 (1956), and private communication.

ratio $\sigma_{p, pn}/\sigma_{p, 2p}$; this is not compatible with a pure evaporation mechanism. An excited Ce¹⁴² nucleus is, as shown in Table D, at least 2.4 to 4.4 times as likely to evaporate a neutron as a proton, the value depending on the excitation energy. Actually these numbers are probably lower limits for the ratio of evaporation
widths. As shown by Eisberg et al.,¹⁸ (α, ϕ) reactions i widths. As shown by Eisberg et al ,¹⁸ (α, p) reactions in this energy range appear to take place partially by a direct interaction mechanism; the same is presumably true for (α,n) reactions to roughly the same extent, so that after subtraction of these contributions the ratio of evaporation-controlled (α, n) and (α, p) cross sections would presumably be greater than the experimentally measured ratio. Furthermore, because of the high Coulomb barrier for α particles, the cross-section ratio could not be measured at sufficiently low energies to exclude some competition of the $(\alpha, 2n)$ with the (α, n) reaction. According to Wapstra's mass tables,¹⁹ the β reaction. According to Wapstra's mass tables,¹⁹ the Q values for (α,n) , (α,β) , $(\alpha,2n)$, and (α,pn) reactions on Ba¹³⁸ are 7.6, 9.2, 13.3, and 16.3 Mev, respectively. Thus, even at 19-Mev bombarding energy, (α, n) might be depleted somewhat by $(\alpha,2n)$, whereas (α,pn) or $(\alpha, n\rho)$ can hardly be significant because protons must be emitted with a few Mev kinetic energy. The effect of this competition from $(\alpha, 2n)$ processes cannot be large up to \sim 25 Mev; otherwise the sum of the (α, β) and (α, n) cross sections could not be so nearly equal to the total α -particle capture cross section at 25 Mev (see Fig. 2), which was computed according to continuum theory²⁰ for a radius parameter $r_0 = 1.5 \times 10^{-13}$ cm.

Comparison with the Ba¹³⁸ (α,n) and Ba¹³⁰ (α,ρ) reactions makes it thus appear that the (p, pn) and $(p, 2p)$ reactions of Ce¹⁴² do not, at 0.4 Bev, predominantly proceed through evaporation from a Ce¹⁴² compound nucleus. The value for the ratio $\sigma_{p, pn}/\sigma_{p, 2p}$ which would be expected from a pure knock-on mechanism may be estimated from the recent Monte Carlo calculations of estimated from the recent Monte Carlo calculations of
intranuclear cascades by Metropolis *et al.*²¹ These authors investigated 796 cascades initiated by 460-Mev protons incident on Ce¹⁴⁰, and found 8 cascades leading to Ce¹³⁹ and 6 leading to La¹³⁹ with residual excitation less than 10 Mev. These may be considered the (p, pn) and $(p, 2p)$ knock-on products, respectively. Thus the calculation gives for pure knock-on production a value of $(8\pm 2.8)/(6\pm 2.4)=1.3\pm 0.7$ for the ratio $\sigma_{p,~p}(\sigma_{p,~2p}$. However, the Monte Carlo calculations do not support a pure knock-on mechanism. Some cascades were found to result in Ce¹⁴⁰ and Pr¹⁴⁰ nuclei with excitations that might result in evaporation to Ce¹³⁹ and La¹³⁹. The probability that any given one of these excited nuclei would indeed result in Ce¹³⁹ or La¹³⁹ as an end product was estimated for the particular excitation energy involved from the experimental data for (α,n) and (α, ρ) reactions on Ba¹³⁸ (see Fig. 2). Thus, in addition to the knock-on (p, pn) and $(p, 2p)$ products (8 and 6, respectively), there are 7.8 (p, pn) and 1.9 $(p, 2p)$ products by an evaporation mechanism. The over-all calculated $\sigma_{p, pn}/\sigma_{p, 2p}$ ratio is $(15.8 \pm 4.0)/(7.9 \pm 2.8)$ $=2.0\pm0.9$, which agrees within the errors with the experimental ratio of 1.58 ± 0.19 measured at 0.4 Bev. Thus, with the aid of the cascade calculations, one may conclude that, at this bombarding energy, (p, pn) and $(p, 2p)$ reactions proceed by a mixture of pure knock-on and knock-on followed by evaporation, with the latter mechanism contributing of the order of 50% of the (p, pn) and 25% of the $(p, 2p)$ cross section.

Whereas the experimentally determined ratio of $\sigma_{p, pn}$ to $\sigma_{p, 2p}$ is thus reasonably well reproduced by the cascade calculations of Metropolis *et al.*,²¹ the magnicascade calculations of Metropolis et al.,²¹ the magni tudes of the cross sections are not. The calculations predict $\sigma_{p, pn} = 28 \pm 7$ mb and $\sigma_{p, 2p} = 14 \pm 5$ mb, to be compared with the experimental values of 86.2 ± 1.6 mb and 54.4 ± 4.6 mb, respectively. This underestimate of (p, pn) and $(p, 2p)$ cross sections by factors of about 3 is a general feature of the calculations by Metropolis $et \ al.^{21}$; as already pointed out by these authors, it may be the result of the neglect of a diffuse nuclear boundary in the model used for the calculations. Whether refinements in the model would also change the predicted relative contributions of knock-on and evaporation mechanisms to the cross sections is not clear.

Energy Deyendence

As may be seen from Table I and Fig. 1, both the $Ce^{142}(p, pn)$ and $Ce^{142}(p, 2p)$ cross sections drop sharply (by factors of 2.8 ± 0.2 and 3.5 ± 0.7 , respectively) as the proton energy is raised from 0.4 to 1.0 Bev. With further increases in incident energy to 3 Bev, the (p, pn) cross section stays almost constant, whereas the $(p, 2p)$ cross section decreases by another factor of 3.7 ± 0.7 . In other words, the ratio $\sigma_{p, p} n / \sigma_{p, 2p}$ increase monotonically with energy in the energy range investigated.

Qualitatively, it is easy to see why the (p, pn) and $(p, 2p)$ cross sections should decrease as the incident energy is raised. Although the total elementary $p-\phi$ and $n-p$ collision cross sections increase with increasing energy $n-p$ collision cross sections increase with increasing energy
above 400 Mev,²² this increase is due to the onset of pion production, and the elastic $p-p$ cross section has actually been found to decrease^{23,21}; the elastic $p-n$ energy is raised. Although the total elementary p - p and n - p collision cross sections increase with increasing energy above 400 Mev,²² this increase is due to the onset of pion production, and the elastic p - p of the increased total nucleon-nucleon cross sections, the mean free path of a proton in nuclear matter becomes smaller and therefore the probability that an incident proton makes one and only one collision in

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¹⁸ Eisberg, Igo, and Wegner, Phys. Rev. 100, 1309 (1955).
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traversing a Ce nucleus decreases. Furthermore, a pure knock-on mechanism for (p, pn) and $(p, 2p)$ reactions requires in general an elastic encounter, and thus a decrease in the elastic nucleon-nucleon cross sections would further lower the (p, pn) and $(p, 2p)$ cross sections. Inelastic nucleon-nucleon collisions (e.g., $p+p\rightarrow p+n+\pi^+, p+n\rightarrow p+n+\pi^0, p+n\rightarrow p+p+\pi^$ $p+p\rightarrow p+n+\pi^+$, $p+n\rightarrow p+n+\pi^0$, $p+n\rightarrow p+p+\pi^-$,
etc.) can lead to "p, pn" or "p, 2p" products only if all three collision products escape from the nucleus without depositing more than \sim 10 Mev of excitation. This is very unlikely, especially since the pions have large scattering cross sections.

It is not so easy to rationalize the observed increase in the ratio $\sigma_{p, \, pn}/\sigma_{p, \, 2p}$ with increasing proton energy. One might try to interpret this increase in terms of elementary cross sections by postulating that the ratio of elastic to total cross section decreases more rapidly with increasing energy for $p\n-*p*$ than for $n\n-*p*$ collisions. This effect is qualitatively very reasonable, since some $n-p$ interactions take place in the isotopic spin state $T=0$ whereas pion production presumably occurs primarily²⁵ for $T=1$. However, this explanation does not appear to be sufficient to account quantitatively for the observed results. Measurements of elastic p - p for the observed results. Measurements of elastic p - p scattering at 2.0,²⁶ 2.2,²⁴ and 4.4²⁴ Bev indicate ratios of elastic to total cross section no less than 0.3. Thus, since the total $p-p$ and $p-n$ cross sections are about equal in this energy range, one would have to assume no pion production at all in $n-p$ collisions to come even close to the experimental $\sigma_{p, pn}/\sigma_{p, 2p}$ ratio. This is, of course, unreasonable. Alternatively one has to postulate that processes other than simple elastic collisions contribute to the observed reactions, especially the (p, pn) reaction, at the higher energies. The situation then becomes much too complex for even qualitative reasoning without detailed computations.

For a more quantitative comparison with the cascade-evaporation model of nuclear reactions one
may turn again to the calculations by Metropolis *et al.*²¹ may turn again to the calculations by Metropolis $et al.²¹$ which include a study of 563 cascades initiated by 1.8-Bev protons incident on Ce^{140} . The results can be compared with the 2.2-8ev data; however the comparison suffers from very poor statistical accuracy due to the small number of calculated events. With the same criteria used as for the 0.46-Bev cascades one finds 3 and 1 cascades, respectively, leading to (p, pn) and $(p, 2p)$ by pure knock-on, one of the (p, pn) cascades being accompanied by π^0 emission. In addition there is one cascade among the 563 studied which leads to Ce^{140} at about 30-Mev excitation and would thus contribute \sim 0.6 Ce¹³⁹ nucleus and \sim 0.2 La¹³⁹ nucleus by evaporation. Thus the computed ratio is $\sigma_{p, pn}/\sigma_{p, 2p} = (3.6$ \pm 1.9)/(1.2 \pm 1.1)=3 \pm 3, a result which is, of course, not very meaningful although compatible with the experimental value of 5.0 ± 0.6 found at 2.2 Bev. The absolute values of the cross sections deduced from the cascade calculations are $\sigma_{p, pn} = 9 \pm 5$ mb and $\sigma_{p, 2p} = 3$ ± 3 mb, again a factor of 2 or 3 smaller than the experimental values. Within the rather large errors, the energy dependence of the cross section appears to be reasonably well reproduced by the Monte Carlo
calculations.²¹ calculations.

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²⁵ See, e.g., Dzhelepov, Satarov, and Golovin, Doklady Akad.
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²⁶ Barge, Barton, and Smith (private communication).