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Survey of (*p*,*n*) Reactions at 12 Mev

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Results of measurements of twenty (*p*,*n*) cross sections on elements from atomic number 21 to 58 are given. The data indicate total reaction cross sections which, when interpreted in terms of a totally black square well potential of depth 20 Mev, correspond to a potential radius of 1.55 to $1.65A^{\frac{1}{3}}\times 10^{-13}$ cm.

INTRODUCTION

TOTAL reaction cross sections of nuclei are currently of interest since they should yield an experimental check of the depth and extension of the imaginary potential employed in the optical model of the nucleus.[1,2] Optical model calculations usually assume the imaginary potential to be of the same form as the real potential but there appears to be no reason for disallowing entirely different shapes and extensions. In view of the striking successes of the optical model in the description of elastic nuclear events, the validity of its description of absorptive processes will be of considerable interest.

Experimental determination of total reaction cross sections is, however, not as direct as one might desire. In charged particle bombardments at medium energies ($\approx$20 Mev) a number of types of reaction are possible, and usually, to obtain the total reaction cross section, it is necessary to measure *all* the individual reactions; this is often not possible, and almost never easy. As the bombardment energy is lowered the (*x*,*n*) reaction becomes the chief contributor and so measurement is facilitated. On the other hand, if one goes too low in energy, for charged particles, barrier penetration corrections in the theoretical interpretation of the data become large and hence the possibility of large errors in interpretation are introduced. Measurements of total reaction cross sections by neutron bombardment, at these energies, involve (*n*,*n*′) in a prominent way and hence are at this time of great experimental difficulty.

In the present experiment it was attempted to select the bombarding energy and the range of atomic numbers such that a reliable and easily interpretable indication of the magnitude of the total reaction cross section could be obtained with minimum experimental effort. The bombarding energy of 12 Mev is high enough to be above the Coulomb barrier for all nuclei in the survey (thus reducing barrier penetration effects) while at the same time being low enough so that almost all reactions are (*p*,*n*) and hence determination of the total reaction cross section involves the minimum number of experimental measurements. In the present paper the cross sections are interpretated in terms of the radius of totally black square well potential[3] which would give the same cross section since total reaction cross sections calculated on the basis of the optical model are not as yet available. It is hoped that such theoretical optical model calculations will soon become available and that the data described herein will constitute a reliable experimental basis for comparison.

EXPERIMENTAL METHOD

Cross-section measurements were performed by activation methods in the internal circulating beam of the ORNL 86-inch cyclotron. Targets were bombarded for a short period, the induced activity was observed in a counter, the product nuclei from the reaction were identified by means of their radioactive half-lives, and cross sections were computed from the intensity of the particular half-life. The proton beam current was measured by means of a comparison technique in which the activity induced by the reaction being investigated was compared with the Zn^{63} activity induced in a sample of copper exposed during the same bombardment. The comparison method has the advantage that all energy independent beta-counting errors cancel, provided the samples to be compared are counted in identical geometrical and physical conditions. This stipulation was rigidly adhered to during the course of the experiment. The comparison method necessarily implies that all cross sections obtained are relative and some additional measurement must be relied on for an absolute scale. For this purpose the measurement by Ghoshal[4] of the $Cu^{63}(p,n)Zn^{63}$ cross section at 12 Mev is ideal. Any error in his value of 530 mb would be reflected as an error in the scale factor of all the measurements, whereas relative values would be unaffected.

Target materials for the bombardments were in some cases the pure element and in other cases the chemical compounds, as follows: $Sc_2(SO_4)_3$, CrF_2, Ni, Zn, GaF_3, NaBr, RbCl, Y_2O_3,[5] $Zr(NO_3)_4$, Ru, Cd, $CsNO_3$, Ce_2O_3, Cu, and CuO. For the measurements on nickel, zinc, and cadmium, thin metal foils were used ($\approx$20 mg/cm^2), whereas all other materials were in the form of a fine powder. The copper monitor was introduced, in the case of the metal foil targets, by stacking the

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[1] D. S. Saxon and R. D. Wood, Phys. Rev. **95**, 587 (1954).

[2] Eisberg, Gugelot, and Porter, Editors, Brookhaven National Laboratory Report, BNL-331, 1955 (unpublished).

[3] M. M. Shapiro, Phys. Rev. **90**, 171 (1953).

[4] S. N. Ghoshal, Phys. Rev. **80**, 939 (1950).

[5] We are indebted to G. E. Boyd of the Chemistry Division of Oak Ridge National Laboratory for the 99% pure Y_2O_3.

target foil along with a 16-mg/cm^2 copper foil, in a thick window frame so that the same proton current passed through both foils. Where targets consisted of powdered materials, copper was introduced in the form of powdered copper oxide which was thoroughly mixed with the target material by a grinding process. Any imperfection in the mixing would result in random variance from one measurement to the next and hence would be reflected as an increased standard deviation in the results from various measurements.

After bombardment, the targets were removed from the cyclotron, and the radioactivity induced by various reactions was observed under end-window Geiger counters enclosed in standard aluminum-lined lead castles. Decay curves were plotted and analyzed into their constituent half-lives; the half-lives were then used to identify the nuclides from which the various radiations emanated. In every case, the production of a particular activity by reactions other than the one being studied was prohibited by application of the energy conservation theorem, so that the intensity of a particular half-life could be straightforwardly converted into a cross section by using known decay scheme information.

Most samples were counted on thick backings so that saturation back scattering was obtained. Under these conditions the number of back-scattered betas has been found by Yaffe[6] to be essentially independent of beta energy over the range of beta energies which were encountered. A few samples were counted on nonthick backings in cases where the energies to be compared were sufficiently similar to make substantial error unlikely. Corrections for absorption in the air and in the counter windows were made in the usual way by extrapolating an experimental absorption curve back to zero absorber. Correction for self-scattering and self-absorption in the samples were made experimentally at two energies and reasonable agreement with the results of Nervick and Stevenson[7] was obtained. The latter results were, therefore, assumed to be adequate for making corrections in data involving other beta energies. In addition to the above corrections, experimental errors were enlarged by from 10 to 20% (depending on the difference in energy of the betas being compared) to take account of possible additional beta-counting errors.

In a few cases activities were determined by counting conversion electrons, where the conversion coefficients were large and well known. In these cases corrections for self-absorption and self-scattering were made by arbitrarily assuming the corrections to be equivalent to those for a beta of twice the energy. Absorption curves taken externally showed the above assumption to hold reasonably well for external absorption over the range of thicknesses encountered experimentally.

Decay scheme information was in general taken from the table of isotopes of Hollander, Perlman, and Seaborg,[8] unless more recent information was available.[9] For the isotopes Sc^{45}, Ni^{60}, Zr^{95}, and Ru^{99}, experimental positron to K-capture ratios were not available, and in these cases the amount of K-capture was estimated from Fermi theory, following the work of Feenberg and Trigg.[10] The third column of Table I shows the percentage of radioactive decays assumed (in the calculation of cross sections) to occur by particle emission for each isotope. Similarly the second column shows the half-life assumed for each radioisotope. No attempt was made to evaluate possible systematic errors caused by faulty decay-scheme information. Every effort was made, however, to hold such errors to a minimum by carefully selecting the best information available. If a better value for any of the entries in columns 2 and 3 of Table I is subsequently found, the observed value of the cross section may be corrected accordingly.

The energy of the proton beam was measured on each cyclotron bombardment by observing the 38-min activity induced in a copper foil stack bombarded behind a thick window frame. The induced activity was then unfolded into an energy spectrum by using the accurately known Cu^{63} excitation function and solving approximately the resulting integral equation by the method of Cohen.[11] The beam energy spectrum was found to have a full width at half-maximum of 1 Mev and the maximum was found to fluctuate from one run to the next over a region of 1.2 Mev with the maxima from 60% of the runs lying within a region having a full width of 0.4 Mev, centered at 11.9 Mev. The 12-Mev energy was obtained from the 86-inch cyclotron by passing the beam through an appropriate copper absorber. An attempt was also made to obtain this energy by operation at reduced radius but was discontinued when the energy spectrum obtained was found to be less satisfactory than that obtained by absorption. All cross sections were measured from four to twelve times on several cyclotron runs so that the values obtained are, hence, average values over the energy distribution of the proton beam.

As an experimental check on the entire method, measurements of known cross sections were made whenever possible and reasonable agreement was obtained.

RESULTS

Table I, column 4 gives the measured values obtained for the various cross sections and column 5 gives the estimate of experimental errors. The errors quoted are the standard deviation for the results from

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[6] L. Yaffe, J. Chem. Soc. S341 (1949).

[7] W. E. Nervick and P. C. Stevenson, University of California Radiation Laboratory Report UCRL-1575, 1951 (unpublished).

[8] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 569 (1953).

[9] Nuclear data accumulation, Nuclear Science Abstracts, 1952–1954.

[10] E. Feenberg and G. Trigg, Revs. Modern Phys. **22**, 339 (1950).

[11] B. L. Cohen, Oak Ridge National Laboratory ORNL-1347, 1952 (unpublished).

TABLE I. Experimental results. Columns 2 and 3 show the assumed half-lives and percent particle emission (taken from references 8, 9, and 10) which were used in the calculation of the cross section; Columns 4 and 5 contain the experimental data; Columns 6 and 7 show energetic thresholds for the (p,n) and $(p,2n)$ reactions (calculated from reference 19).

Reaction	Half-life	Percent $(\beta^+ + \beta^- + e)$	Observed (p,n) cross section (mb)	Error (mb)	Thresholds (Mev)	
					(p,n)	$(p,2n)$
$\mathrm{Sc}^{45}(p,n)\mathrm{Ti}^{45}$	3.08 hr	82.4	350	130	2.73	>12.0
$\mathrm{Cr}^{52}(p,n)\mathrm{Mn}^{52*}$	21 min	99	210	40	5.8	
$\mathrm{Cr}^{52}(p,n)\mathrm{Mn}^{52}$	5.6 days	35	75	15	5.4	>16.0
$\mathrm{Ni}^{60}(p,n)\mathrm{Cu}^{60}$	24.6 min	100	370	65	6.8	>12.3
$\mathrm{Ni}^{61}(p,n)\mathrm{Cu}^{61}$	3.4 hr	68	590	160	2.8	14.6
$\mathrm{Ni}^{64}(p,n)\mathrm{Cu}^{64}$	12.8 hr	58	1210	450	2.4	10.3
$\mathrm{Zn}^{66}(p,n)\mathrm{Ga}^{66}$	9.2 hr	64	585	60	5.9	≈13.7[a]
$\mathrm{Zn}^{68}(p,n)\mathrm{Ga}^{68}$	68 min	85	1100	150	3.6	11.9[a]
$\mathrm{Ga}^{69}(p,n)\mathrm{Ge}^{69}$	39.6 hr	33	960	350	4.0	11.5[a]
$\mathrm{Br}^{79}(p,n)\left\{\begin{matrix}\mathrm{Kr}^{79*}\\ \mathrm{Kr}^{79}\end{matrix}\right.$	34 hr	7.0	1050	350	2.7[a]	10.8
$\mathrm{Rb}^{87}(p,n)\mathrm{Sr}^{87*}$	2.8 hr	22	220	70	2.0	6.5?
$\mathrm{Y}^{89}(p,n)\mathrm{Zr}^{89*}$	4.4 min	7.0	170	60	4.2	
$\mathrm{Y}^{89}(p,n)\left\{\begin{matrix}\mathrm{Zr}^{89*}\\ \mathrm{Zr}^{89}\end{matrix}\right.$	78 hr	24	750	100	3.6	13.5[a]
$\mathrm{Zr}^{90}(p,n)\mathrm{Nb}^{90}$	15 hr	41	1200	250	7	15.4[a]
$\mathrm{Ru}^{99}(p,n)\mathrm{Rh}^{99}$	4.5 hr	11.7	420	120	2.4	14.0?[a]
$\mathrm{Ru}^{101}(p,n)\mathrm{Rh}^{101}$	4.5 days	10	365	100	1.4[a]	11.6?
$\mathrm{Cd}^{111}(p,n)\mathrm{In}^{111}$	2.8 days	16.7	810	120	1.6[a]	11.9
$\mathrm{Cd}^{114}(p,n)\mathrm{In}^{114*}$	50 days	97	305	50	3.0	9.6
$\mathrm{Cs}^{133}(p,n)\mathrm{Ba}^{133*}$	38.8 hr	71	270	60	1.0	7.0?
$\mathrm{Ce}^{142}(p,n)\mathrm{Pr}^{142}$	19.2 hr	100	110	40	3.5	7.0

[a] Beta energy estimated from systematics; K. Way (private communication).

various runs plus a percentage error (mentioned above) allowed for possible unknown counting errors. The results are also plotted in Fig. 1 with the various symbols being defined in the caption. Also included in Fig. 1 are theoretical total reaction cross sections calculated by Shapiro[3] for a totally black square well of radius $R = r_0 A^{\frac{1}{3}} \times 10^{-13}$ cm, where r_0 has the value indicated on each of the curves. The theoretical curves are spread out into bands to correspond to the experimental distribution of proton energies. It can be seen from the width of the bands that the dependence of the total cross section on energy is not critical and hence the experimental spread in energy should not have any important effect on the final results.

Two striking features show up at once in Fig. 1, (a) the large magnitude of the cross sections observed, and (b) the large fluctuation in cross section observed from one nucleus to the next. Considering first the large magnitude of the cross sections, one notes that five of the observed cross sections fall near or above the $r_0 = 1.7$ curve, indicating extremely strong absorption. Unfurtunately four of these large values suffer in experimental accuracy for various inherent reasons.[12] The one remaining very large cross section, Zn^{68}, was, however, amenable to an accurate measurement and, therefore, in order to sharply pin point at least one of the very large cross sections, a number of additional runs were made. As a result the measurement on Zn^{68} has the best relative accuracy in the survey. It is interesting to note that Zn^{68} displayed an r_0 of 1.8[13] at 6.7 Mev and that systematics of measurements of (p,pn) and $(p,2n)$ cross sections at 22 Mev[14] also indicate a large total cross section for this isotope. If one bears in mind that there are a number of possible factors which can cause the (p,n) cross section to be substantially smaller than the total reaction cross section, (but it can in no way become larger), the raw experimental data may be regarded from two points of view. On the one hand the distribution of observed cross sections can be construed as indicating a rising sequence whose upper limit, represented by the large cross sections, indicates the true approximate value of the total reaction cross section for all isotopes. The total reaction cross section, following this picture, would vary only slightly from one

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[12] Available information on the decay scheme of Zr^{90} is very meager, and it was necessary to rely on a theoretical determination of the amount of K-capture. Also, the assumption of an allowed transition may not hold. Br^{79} has the lowest beta energy of any of the isotopes studied in the experiment and hence the amount of K-capture is large. The ratio of K-capture to beta-plus-decay has been carefully measured[8] but because of the small amount of particle emission ($\approx 5\%$), a sizable error could still result in the cross section determination. For Ga^{69} the rather rough experimental determination of the K-capture ratio given in reference 8 was used. This value is not in good agreement with the prediction of Fermi theory, and if the theoretical value were used a much smaller cross section would result. Ni^{64} is a 1.0% isotope and the radioactivity from the product Cu^{64} is hence weak and difficult to observe. The situation is further complicated by the 18-hr (p,α) activity from Ni^{58} which made it necessary to follow decay curves for a very long time in order to accurately separate the 18-hr and 12.8-hr activities. As a check on the adequacy of the decay curve analysis, a chemical separation was made in one run to eliminate the 18-hr activity, and the resulting value fell within the random scatter of determinations from the other runs.

[13] Blaser, Boehm, Marmier, and Scherer, Helv. Phys. Acta. 24, 441 (1951). In this and all following instances where the data of this reference are interpreted in terms of nuclear radii, the radius quoted is that which results from application of the theory of reference 3.

[14] B. L. Cohen and E. Newman, Phys. Rev. **99**, 718 (1955).

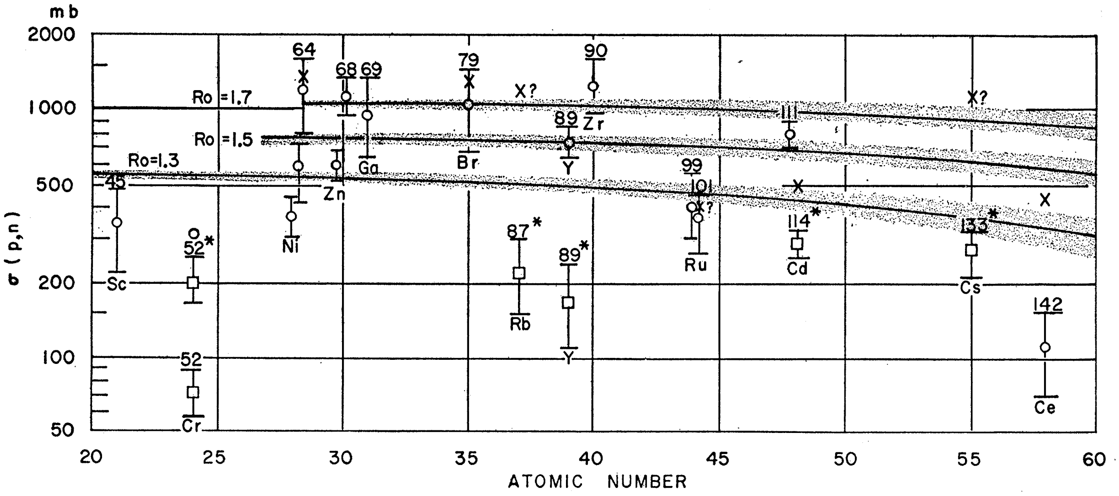

FIG. 1. (*p*,*n*) cross sections in millibarns. ○ —measured total (*p*,*n*) cross section for designated isotope; □—partial (*p*,*n*) cross section leading to a particular isomeric state of the designated isotope; × —observed (*p*,*n*) cross section plus theoretical (*p*,2*n*) cross section estimate. Curves are calculated total reaction cross sections taken from reference 3 for three values of r_0. The dotted bands indicate the spread caused by the distribution of bombarding energy. (Note: "Ro" in three places on the figure should read "r_0.")

isotope to the next in accord with the $A^{\frac{2}{3}}$ law and the small (*p*,*n*) cross sections observed in several cases would be an indication of substantial competition from other reactions. On the other hand, one might assume the observed (*p*,*n*) cross sections to be equivalent indicators of the total reaction cross section in all cases, so that one is left with a total reaction cross section which fluctuates rather violently, which is on the average rather small but still becomes very large in selected cases. The following examination of competing processes indicates the former of the above points of view to be more plausible.

First of all, one notes the trivial case posed by the three isotopes Rb^{87}, Cd^{114}, and Cs^{133} for which it was possible to measure only the partial (*p*,*n*) cross section leading to one of the isomeric states of the product nucleus. The total cross section must therefore be larger by at least the amount of (*p*,*n*) transitions to other isomeric states. In two cases, Cr^{52} and Y^{89}, it was possible to measure the cross section for each isomer, and in neither case was one of the isomers negligible. Thus, for the remaining cases where transitions to isomeric states were possible but unmeasured, it seems quite probable that the total cross section should be raised an appreciable amount.

A second competing process which should be considered is the case wherein the (*p*,*n*) reaction leaves a residual nucleus with sufficient energy for further particle emission, resulting in depletion of the number of residual nuclei and hence in a smaller observed (*p*,*n*) cross section. Thus, one might define a "true" cross section to be $\sigma'(p,n)=\sigma(p,n)+\sigma(p,np)+\sigma(p,2n)$. With respect to the $\sigma(p,np)$ term, the excitation functions observed by Cohen *et al.*[15] for $\sigma(p,np)+\sigma(p,pn)$ indicate reactions of this type to be negligible to about 4 Mev above threshold, in the mass region covered in this experiment, presumably because of the kinetic energy which the proton needs to penetrate the Coulomb barrier. The application of this energetic criterion eliminates $\sigma(p,np)$ for all isotopes except Ni^{61}. For Ni^{61}, $\sigma(p,np)$ was calculated on the basis of statistical theory and found to be considerably less than the experimental error in $\sigma(p,n)$. Hence, for all cases $\sigma(p,np)$ may be neglected. On the other hand, (*p*,2*n*) reactions rise quite rapidly from their energetic thresholds and since, as the sixth column of Table I shows, the (*p*,2*n*) thresholds are in a number of cases below the 12-Mev bombarding energy, one expects a substantial contribution from this reaction. The amount of the (*p*,2*n*) contribution may, however, be quite reasonably estimated from the statistical theory of nuclear reactions, if one knows the (*p*,2*n*) threshold and the energy distribution of the neutrons emitted from the initial compound nucleus. Following the development of Blatt and Weisskopf,[16] one finds

$$\sigma'(p,n)=\sigma(p,n)(1+\Delta E_{2n}/T)^{-1}\exp(\Delta E_{2n}/T),$$

where ΔE_{2n} is the excess of bombarding energy above the (*p*,2*n*) threshold and where T is a nuclear temperature. For this calculation T was estimated by attaining a best fit to several known (*x*,2*n*) *vs* (*x*,*n*) ratios.[17,18] The value obtained in this fashion was $T=1.8$ Mev. With this value of T, the $\sigma'(p,n)$ cross section was calculated for all cases in which the bombarding energy exceeded the (*p*,2*n*) threshold. The resulting "true" (*p*,*n*) cross section estimates are indicated in Fig. 1 by ×. The

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[15] Cohen, Newman, Charpie, and Handley, Phys. Rev. **94**, 620 (1954).

[16] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

[17] Bleuler, Stebbins, and Tendam, Phys. Rev. **90**, 460 (1953).

[18] A. I. Berman and K. L. Brown, Phys. Rev. **96**, 83 (1954).

threshold values used in this calculation were computed from the table of atomic masses of Cushman.[19] For three isotopes, no experimental mass information was available and values from the semiempirical mass formula were used. In these cases the corrected cross sections and thresholds are marked with a question mark to indicate their lack of reliability.

The final substantial competing process which one must consider is the (p,p') reaction. Corrections for this type of competition, made by application of the statistical theory of nuclear reactions,[16] do not substantially reduce the magnitude of the observed cross section fluctuations. Paul and Clarke[20] have observed, however, that for 14-Mev neutron-induced reactions the amount of proton emission can fluctuate by orders of magnitude about the theoretical prediction in the mass region surveyed. At 14 Mev, (n,p) cross sections of 100 to 200 mb were found quite often, and several still larger values were observed. Also, proton emission is energetically favored by an additional 5 Mev on the average, when the reactions are proton-induced, as in this experiment, rather than neutron-induced, as in Paul and Clarke's work. Thus, on the basis of this data (p,p') cross sections varying widely up to several hundred millibarns would seem quite likely, and could adequately account for the observed fluctuations in the (p,n) cross section. If one assumes this explanation of the fluctuations, it then follows, as mentioned before, that the observed (p,n) cross sections are a rising sequence whose upper limit indicates the value of a slowly varying total reaction cross section. The radius of equivalent black square well thus implied is quite large, corresponding to an r_0 of at least 1.65 and more probably 1.7. The evidence for this large value of the total reaction cross section is indirect and hence must be considered as doubtful until direct absolute measurements of (p,p') cross sections at these energies are made. In support of this view, however, one should note the large number of evidences of slowly varying total reaction cross sections[21–24] which would be sharply contradicted if one were to accept the theoretical estimates for (p,p') competition and the sharp fluctuations in total reaction cross sections thus entailed.

On the other hand, in possible support of the alternative picture of a sharply varying total cross section, one should note the many successes of the shell model of the nucleus. On the basis of this theory, nuclear characteristics are in a large measure determined by the behavior of one or a few surface nucleons, and hence qualitatively one might expect rather large fluctuations in the values of properties, such as the total cross section, to result from the addition of a single neutron. Also it should be noted that the evidences mentioned above[22–24] for a relatively constant total cross section are all based on studies of natural elements and hence an averaging over isotopes has taken place, which would tend to mask sharp fluctuations in the total cross section. This masking would be particularly effective if some systematic pattern of deviation existed about particularly stable nuclear configurations, as is perhaps plausible on the basis of the shell model.

The fact that the total cross section does not sharply fluctuate with proton number could be an indication that the nuclear fringe (which essentially determines the cross section) consists primarily of neutrons as indicated by recent work of Hess.[25] Thus, in view of these and other arguments, there appears to be, at present, insufficient evidence for definitely attributing the (p,n) fluctuations to (p,p') competition, since the possibility of large total reaction cross section fluctuations cannot be wholly excluded. Direct measurements of (p,p') total cross sections are needed in order to arrive at a definite conclusion on this point; they are planned as an extension of this survey.

Finally, one should note that the results of the present survey when interpreted in terms of the equivalent black square well, give radii in essential agreement with previous charged particle total reaction cross section determinations.[13,26] The r^0 implied is quite large, being from 1.55 to 1.65 depending on which of the above views of (p,p') competition is assumed. In terms of the optical model these large total reaction cross sections apparently imply an extensive and rather intense imaginary potential. It is hoped that theoretical work will soon indicate whether such strong absorption can be described with the presently assumed values of the optical model parameters.

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[21] Feshbach, Porter, and Weisskopf, Phys. Rev. **96**, 448 (1948).

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[23] N. Nereson and S. Darden, Phys. Rev. **94**, 1678 (1954).

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[26] B. L. Cohen and G. H. McCormick, Phys. Rev. **96**, 722 (1954).