and between the (p,pn) excitation functions[6,19] for Cu^{63} and Cu^{65}.

We might note two other indications that the reactions observed are probably not proton emission following (p,n) reactions. Firstly, Gugelot's measurements[16] indicate that the energy spectra of emitted protons are the same in copper as in nickel and iron. Measurements at higher energies[9] clearly show that the first particle emitted in nickel is a proton, and it seems relatively certain from the preceding discussion that the same is true for iron.

Secondly, a remeasurement was made of the excitation functions in Cu^{63}, and it was found that $[\sigma(p,n)+\sigma(p,2n)+\sigma(p,pn)]/\sigma_p$ decreases by 15% as the energy is increased from 11 to 15 Mev. This indicates that the (p,n) cross section probably drops off from competition with the unobserved (p,p') reaction rather than from competition with (p,np).

The conclusions of this paper that F_p/F_n is abnormally large is also supported by two other experiments:

(1) Meadows,[25] in measuring excitation functions in copper with high-energy protons, found that not only are the (p,pn) cross sections much larger than the ($p,2n$)'s, but ($p,p2n$) and ($p,p3n$) cross sections are much larger than the ($p,3n$)'s and ($p,4n$)'s.

(2) In the following paper,[11] it is shown that ($p,2p$) cross sections are very large in this mass region, and in slightly lighter nuclei, commonly are the most probable of all reactions.

[25] W. Meadows, Phys. Rev. 91, 885 (1953).

In spite of these arguments, it should always be borne in mind, of course, that we are dealing here with a statistical phenomenon; as such, it is subject to wide fluctuations, as can be seen directly from the data. It would be essentially impossible to prove that the effects found here cannot be explained by a combination of these fluctuations, the Wigner effect,[24] large differences between level densities in even-even and odd-odd nuclei, and contributions from (p,d) "pickup" reactions. Certainly any isolated piece of data can easily be explained in that way. However, after prolonged consideration of the various aspects of the problem, the authors have reached the opinion that the large ratio of proton to neutron emission cannot be explained by the usual nuclear reaction theories. Experiments are being undertaken to further study the problem by observing energy distributions and angular distributions of the emitted protons as a function of bombarding energy.

Before concluding, it is interesting to note that the large (p,pn) cross sections indicate that the neutron energy spectra measured by Gugelot[12] are greatly distorted by neutrons from those reactions.

ACKNOWLEDGMENTS

The authors are greatly indebted to Professor E. P. Wigner and Professor V. F. Weisskopf for their very valuable suggestions, to R. S. Livingston for his continual encouragement, and to J. A. Martin for his stimulating discussions. The As(p,pn) cross section was measured by A. Timnick.

PHYSICAL REVIEW VOLUME 99, NUMBER 3 AUGUST 1, 1955

(p,pn)+($p,2n$) and ($p,2p$) Cross Sections in Medium Weight Elements*

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(Received March 7, 1955)

Several (p,pn)+($p,2n$) and ($p,2p$) cross sections of nuclei of mass 19 to 69 were measured with 21.5-Mev incident protons. For all elements with mass less than 55 and for Ni^{58}, the (p,pn)+($p,2n$) cross section is very much less than the total reaction cross section. Detailed analysis of the data indicates that this is largely due to competition from ($p,2p$) reactions. The conclusion from the previous paper that the ratio of probabilities for proton and neutron emission is much larger than expected seems to be confirmed and extended to lighter nuclei.

INTRODUCTION

In planning our recent survey of activation cross sections for various types of nuclear reactions in medium weight elements, little attention was at first given to the relatively large number of cases where the sum of the (p,pn)+($p,2n$) cross sections can be conveniently measured. It was assumed that, apart from small corrections, these would add up to the total reaction cross section (σ_R).[1] It was soon found, however, that this was not by any means the case. In many of the early measurements, values very much less than σ_R were obtained, and as the data were extended, it turned

* The "(p,pn)+($p,2n$) cross section" is used here to mean the sum of the ($p,2n$), (p,pn), (p,np) and (p,d) cross sections, all of which lead to the same radioactive nucleus (after a beta decay in the first case).

† Present address: U. S. Army.

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

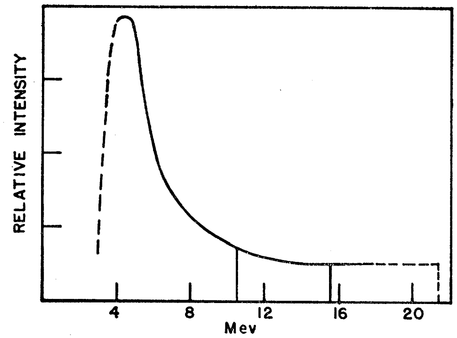

FIG. 1. Assumed energy distribution for 21.5-Mev protons inelastically scattered by copper. This was obtained from reference 3 by assuming the low-energy portion to be unchanged when the incident proton energy is raised from 18 to 21.5 Mev, and smoothly extrapolating the high-energy portion after averaging over resonances. In reference 3, similar results were obtained for iron and nickel.

out that this was true in general for every measurable case of mass less than 55, and for Ni^{58}. A detailed theoretical analysis revealed that competition from $(p,2p)$ reactions might be an important factor in the problem, so that the experiments were extended to include cross section measurements for those reactions wherever possible.

THEORY

In accordance with nuclear reaction theory,[1] a (p,p) reaction generally becomes a (p,pn) reaction when the proton is emitted with low enough energy to leave neutron emission energetically possible. In cases where the residual energy is such that emission of a second proton is possible but neutron emission is not, a $(p,2p)$ reaction should result. When either can be emitted, the Coulomb barrier should suppress proton emission leaving neutron emission more probable, unless this effect is compensated by the extra energy available for proton emission. In order to understand these cross sections we must therefore have some knowledge of the energy distribution with which neutrons and protons are emitted from (p,n) and (p,p) reactions. Measurements of these have been reported by Gugelot[2,3] for 16- and 18-Mev protons. It is assumed here that the low-energy portions of the spectra are not changed by increasing the incident proton energy to 21.5 Mev, and that the high-energy portions extrapolate smoothly to 21.5 Mev after averaging over resonances (and, of course, neglecting the elastic peak in the proton spectra). The proton spectra for copper and aluminum obtained in this way are shown in Figs. 1 and 2. Figure 2 will be used in the analyses for the light elements ($A \leqslant 39$), and Fig. 1 will be used for the heavier elements.

The probabilities for secondary reactions can be seen from Fig. 1. The thresholds for (p,pn) and $(p,2p)$ reactions in copper are about 11 Mev and 6 Mev, respectively. Emission of a neutron following a (p,p) reaction will then be energetically possible if the proton is emitted with an energy less than 11 Mev below the maximum energy, or 10.5 Mev. The probability for this is therefore nearly equal to the area under the curve to the left of 10.5 Mev, divided by the total area under the curve; this is about 72%. If the proton is emitted with an energy between 10.5 and 15.5 Mev, emission of a second proton is energetically possible while emission of a neutron is not, so that a $(p,2p)$ reaction will occur (neglecting competition with alpha emission and electromagnetic de-excitation). The area under the curve between 10.5 and 15.5 Mev is about 13% of the total area, so that at least 13% of (p,p) reactions become $(p,2p)$. However, among the 72% where neutron emission is possible, proton emission will be energetically favored by 5 Mev and may therefore still compete effectively. Thus, of the total number of original (p,p) reactions, somewhat more than 13% become $(p,2p)$, somewhat less than 72% become (p,pn), and 15% remain (p,p). These results are, of course, sensitive to the energetic thresholds for the various reactions. For example, if the $(p,2p)$ threshold is higher than the (p,pn), the former reaction should occur only very rarely.

If the original reaction is a (p,n), an analysis similar to that above reveals that over 90% of the time, a $(p,2n)$ or a (p,np) reaction occurs A measurement of $(p,pn)+(p,2n)$ cross sections should therefore give the sum of nearly all of the original (p,n) cross section, plus much of the (p,p) cross section.

The only other reaction which can occur with appreciable probability is the (p,α) which at this energy is largely $(p,\alpha n)$ and $(p,\alpha p)$. This has been found[4] to have a cross section of about 150 mb or less in this mass region. Therefore, the sum of the $(p,pn)+(p,2n)$ and the $(p,2p)$ cross sections should add up to a large fraction of the total reaction cross section. If the predominant original reaction is (p,n), this must be true of the $(p,pn)+(p,2n)$ alone.

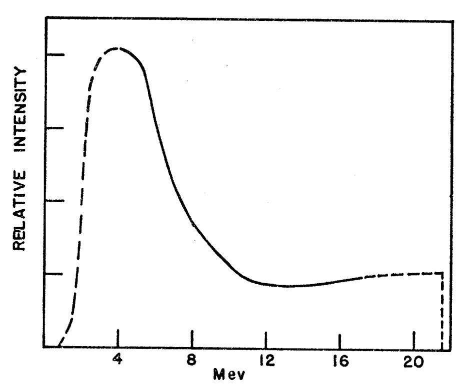

FIG. 2. Assumed energy distribution for 21.5-Mev protons inelastically scattered by aluminum. See caption for Fig. 1.

[2] P. C. Gugelot, Phys. Rev. 81, 51 (1951).
[3] P. C. Gugelot, Phys. Rev. 93, 425 (1954).
[4] D. J. Coombe (private communication).

For the lighter elements, the situation is somewhat different. Figure 2 indicates that an especially large number of high-energy protons are emitted, so that about 25% of the (p,p) reactions are not followed by further particle emission. In addition, the area where (p,$2p$) reactions occur while (p,pn)'s are energetically forbidden is quite appreciable, making up about 25% of the total. Furthermore, Coulomb effects are not as strong here, so that (p,$2p$) also competes effectively with (p,pn) in the remaining 50% of the cases where (p,pn) reactions are energetically possible. Thus, we might expect the total reaction cross sections, after the (p,α) and (p,n) contributions have been subtracted, to consist of about 25% (p,p), considerably more than 25% (p,$2p$), and considerably less than 50% (p,pn).

EXPERIMENTAL

Cross sections for several (p,pn)+(p,$2n$) and (p,$2p$) reactions were measured with the internal, circulating 22-Mev proton beam from the ORNL 86-inch cyclotron. The methods have been described previously.[5] In all of the (p,$2p$) reactions except those on Mg^{25} and Si^{29}, chemical separations were used. For Zn^{68}(p,$2p$)Cu^{67} and for Si^{29}(p,$2p$)Al^{28}, it was found necessary to use separated isotopes.[6] The absorption corrections for the Cu^{67} activity were found to be very large—in fact, much larger than expected even from the very low beta energies involved—so that the uncertainty in that cross section is much larger than in the others. The end product of the reaction Ca^{43}(p,$2p$)K^{42} can also be produced by a (p,He^3) reaction on the fifteen times more abundant isotope Ca^{44}, but the cross section for that has been measured[7] and found to be very small. A small correction has been applied.

The cross section for Ni^{58}(p,$2p$)Co^{57} was measured relative to that for Ni^{58}{(p,pn)+(p,$2n$)}Ni^{57} by radiochemically separating the nickel and cobalt fractions shortly after the bombardment and allowing 36-hour Ni^{57} to decay into Co^{57}. The relative amounts of Co^{57} in the two fractions was then determined by counting on the 130-kev gamma peak with a scintillation spectrometer.

The average error in these cross sections is about 7% as judged by reproducibility, but if one includes beta counting corrections, is probably about 20%. This does not include a possible error of about 15% in the absolute calibration of all cross sections.

RESULTS

The measured cross sections are listed in Table I. The Cr^{52}, Mn^{55}, Fe^{56}, Co^{59}, Cu^{63}, Cu^{65}, Zn^{68}, and Ga^{69} cross sections were taken from reference 5. In all other cases, the end product of the (p,$2n$) reaction was allowed to decay into the same isotope as is produced by the (p,pn) reaction, and only the cross section for production of that isotope was measured. Table I also lists the energetic thresholds for the various reactions.

In several cases, the activity of the end product of the (p,$2n$) reaction has never been detected, so that there exists the possibility that its half-life may be very long rather than very short. In all of these cases but one (Zn^{64}), the ground-state mass can be calculated by assuming that the difference in mass between a nucleus with n protons and (n+2) neutrons, and a nucleus with (n+2) protons and n neutrons is due only to the Coulomb repulsion energy of the two additional protons in the latter. This method has been found to be accurate in explaining known masses, and successfully predicted the maximum beta energy of Ne^{18} before that isotope was discovered.[8] The results of this calculation indicate that in every case, the unobserved activity should be short lived. As a check, a search was made for possible long-lived activities in several of these isotopes. If the half-lives are assumed to be longer than two minutes and to be produced with cross sections equal to the (p,pn) cross sections, the following minimum half-lives are established[9]: Mg^{22}—10^7 years, A^{34}—10^5 years, Ca^{38}—

TABLE I. Summary of measured cross sections and energetic thresholds for various reactions.

Elements	Measured cross sections (mb) (p,pn)+(p,$2n$)	(p,$2p$)	Thresholds (Mev) (p,pn)	(p,$2p$)	(p,$2n$)	(p,n)
$_9F^{19}$	175	···	10.4	8.0	15.3	3.9
$_{11}Na^{23}$	225	···	12.7	9.1	17.8	4.8
$_{12}Mg^{25}$	···	54	7.6	12.6	$\sim$12	5.0
$_{14}Si^{29}$	···	110	8.7	11.8	$\sim$17	5.3
$_{15}P^{31}$	240	···	12.8	7.4	17.8	6.0
$_{17}Cl^{35}$	120[a]	···	13.6	6.6	18.6	6.3
$_{19}K^{39}$	105[a]	···	13.5	6.6	20.1	7.0
$_{20}Ca^{43}$	···	15	8.0	10.9	$\sim$13	3.8
$_{20}Ca^{44}$	···	5.0	11.6	12.7	14.5	4.4
$_{22}Ti^{46}$	<500[b]	···	15.2	11.4	22.6	$\sim$5
$_{24}Cr^{52}$	580	···	12.0	10.9	16.0	5.4
$_{25}Mn^{55}$	>620	···	10.3	8.2	10.4	0.9
$_{26}Fe^{54}$	165	···	14.1	9.7	22.4	$\sim$7.5
$_{26}Fe^{56}$	870	···	11.5	10.6	15.7	5.3
$_{26}Fe^{57}$	···	12.5	7.6	10.5	13.0	2.1
$_{27}Co^{59}$	>540	···	10.9	7.9	10.8	$\sim$1.0
$_{28}Ni^{58}$	240	680	11.9	8.0	20.6	$\sim$9
$_{29}Cu^{63}$	700	···	11.0	5.3	13.4	4.2
$_{29}Cu^{65}$	>500	···	10.4	6.9	10.5	2.1
$_{30}Zn^{64}$	675	···	12.0	7.7	$\sim$18	$\sim$6
$_{30}Zn^{66}$	920	···	10.8	8.7	$\sim$15	5.9
$_{30}Zn^{68}$	>780	3.8	8.9	8.7	10.7	3.6
$_{31}Ga^{69}$	730	···	10.4	6.8	11.0	4.1

[a] The cross sections observed were for the 34-minute and 7-minute activities for Cl^{34} and K^{38}, respectively. Recently, short-lived positron emitting isomers of these nuclei have been discovered, so that the values listed should be considered minima. The actual cross sections are probably about twice as large.

[b] The observed cross section on Ti^{46} includes the cross section for (p,α) reactions on two equally abundant isotopes and a (p,αn) reaction on a much more abundant isotope. They produce activities with about the half-life as that produced by (p,pn)+(p,$2n$) on Ti^{46}.

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

[6] Separated stable isotopes were obtained from the Stable Isotopes Division, Oak Ridge National Laboratory.

[7] H. G. Blosser (private communication).

[8] J. D. Gow and L. Alvarez, Phys. Rev. **94**, 365 (1954).

[9] This work was done in collaboration with W. H. Jones, Oak Ridge Institute for Nuclear Studies research participant from Emory University.

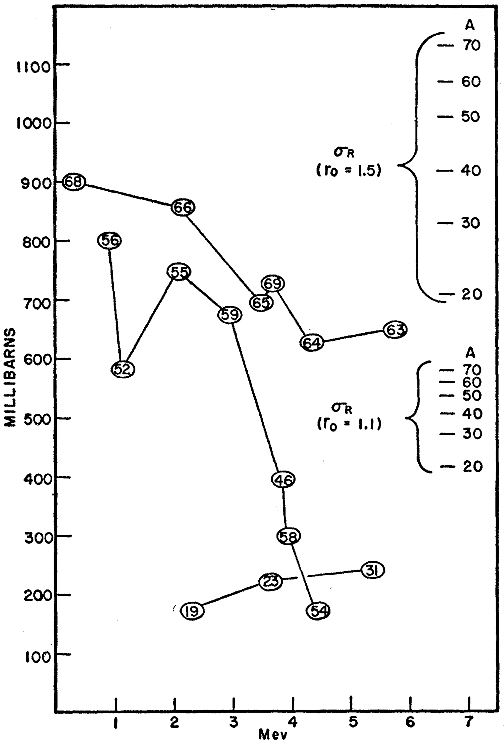

FIG. 3. $(p,pn)+(p,2n)$ cross sections *vs* excess energy available for $(p,2p)$ over (p,pn) reactions. Figures are mass numbers of target nuclei. Lines on right margin are theoretical total reaction cross sections for various masses and constants in the nuclear radius formula.

10^4 years. It therefore seems quite certain that the unknown activities actually are short lived.

CONCLUSIONS

In all six cases where the $(p,2p)$ threshold is higher than the (p,pn), the $(p,2p)$ cross sections from Table I are found to be relatively small, in agreement with the theory. The $(p,2p)$ cross section for Ni^{58} will be discussed below.

Figure 3 shows the $(p,pn)+(p,2n)$ cross sections from Table I plotted *vs* the energy by which the $(p,2p)$ energetic threshold is lower than that for (p,pn). In cases where only minimum values are listed in Table I, the extrapolation is based on the results of reference 5. In no case does this introduce an uncertainty greater than about 15 percent in the points on Fig. 3; this cannot affect the qualitative conclusions. Lines are shown connecting points with mass less than 40, with mass between 40 and 60, and with mass larger than 60. Figure 3 also shows σ_R for various masses and nuclear radii.

When one considers that the observed cross sections should be considerably smaller than σ_R, it is quite apparent (and will become more so when the $(p,2p)$ cross sections are considered below) that nuclear radii corresponding to r_0 equal to at least 1.5 are suggested from the heavy element data. Some of the advantages of determining the nuclear radius by the total reaction cross section have been discussed in a previous paper.[10]

It has been shown previously[5] that for most of the elements heavier than mass 52, (p,n) reactions [ordinarily detected as $(p,2n)$] are only minor contributors to the total reaction cross sections. It is immediately obvious from Fig. 3 that this is also true of all elements lighter than mass 52, and for masses 54 and 58 which were not covered in reference 5. This will be discussed further below.

In view of this result from reference 5, the data for the elements of mass 63–69 in Fig. 3 are in general agreement with the theoretical predictions. Some estimate of the effectiveness of competition from $(p,2p)$ reactions can be made by assuming that it is this competition which causes the lines through the data to slope downward to the right. By extrapolating the lines to zero abscissa, the value for the $(p,pn)+(p,2n)$ cross section, free of competition from $(p,2p)$, is obtained. For masses between 63 and 69, this is between 900 and 1000 mb. It therefore seems that the $(p,2p)$ cross sections for Cu^{63}, Cu^{65}, Zn^{64}, and Ga^{69} are about 200–300 mb. This represents about 20% of $\sigma_R-\sigma(p,\alpha)$, which is in agreement with the prediction of somewhat more than 13%.

The data for the elements of mass 19–39 are not as simple to analyze. The (p,α) cross sections make up an

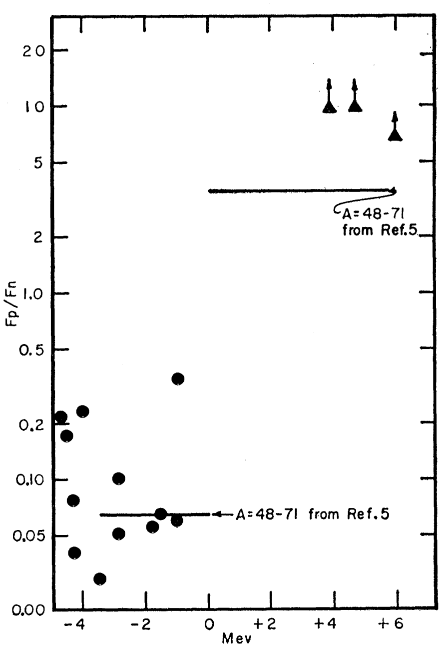

FIG. 4. Ratio of probabilities for emission of protons and neutrons *vs* difference in maximum energy available to them. Triangles are from 21.5-Mev proton data and circles are from 14-Mev neutron data, all in the mass range 16 to 41. Lines are from similar data in the mass range 48 to 71 from Fig. 2 of reference 5.

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

appreciable fraction of σ_R, and they probably vary considerably from element to element. They should be especially large for F^{19} and Na^{23} because they lead to very high energy releases for these isotopes. It is, therefore, difficult to extrapolate the line through the data to zero abscissa. The data certainly agree, however, with the theoretical prediction that $\sigma(p,pn)$ is considerably less than 50% of $\sigma_R-\sigma(p,\alpha)$. For F^{19}, Na^{23}, and P^{31}, it may be estimated as about 30% of this difference.

This data for the light elements can be used to extend the thesis of reference 5 that proton emission is much more probable in 21-Mev proton-induced reactions than in 14-Mev neutron-induced reactions. In view of the very large $(p,2p)$ cross sections, a considerable fraction ($\gtrsim\frac{2}{3}$) of the activity produced by $(p,pn)+(p,2n)$ reactions must be due to (p,pn) rather than to (p,np) or $(p,2n)$. Figure 4 shows a plot of F_p/F_n, the relative probabilities for emission of protons and neutrons, in the original reaction, plotted *vs* the difference in energy available for proton and neutron emission. The 14-Mev neutron data from the (n,p) cross section measurements of Paul and Clarke[11] are also shown. Figure 2 of reference 5 is a similar plot of data for the region of masses 48–71; the lines through the data from that figure are also shown in Fig. 4. It seems quite clear that the conclusion of reference 5—that even after correcting for energetic differences, F_p/F_n is considerably larger at 21.5 Mev than at 14 Mev—is also valid in the lighter mass region.

It should be noted that the very small $(p,pn)+(p,2n)$ cross sections found in the light elements are in agreement with neutron yield measurements.[12] For 23-Mev protons bombarding thick targets of both magnesium and aluminum, only about one neutron was observed for every four nuclear reactions.

For the elements between mass 46 and 59, one would expect cross sections similar to those found in the heavier elements since the Coulomb barriers and energy distributions of emitted particles[2,3] are about the same for the two groups. This expectation is reasonably well fulfilled for Cr^{52}, Mn^{55}, Fe^{56}, and Co^{59}, and the somewhat lower value for Ti^{46} might be explained by the facts that it is the lightest isotope of the group and has a very high (p,pn) threshold. For Fe^{54} and Ni^{58}, on the other hand, the observed cross sections are very much lower than can be easily explained. Fortunately, the $(p,2p)$ cross section in Ni^{58} is measurable (see Table I),[13] so that it is definitely established that the low value of the $(p,pn)+(p,2n)$ cross section is due to competition with the $(p,2p)$. This cannot be explained by a difference in the energy spectrum of the emitted particles (these were measured in references 2 and 3). One possibility that perhaps should not be overlooked is that Ni^{57} and Fe^{53} may have short-lived isomers which decay by positron emission. This could also explain why the activation cross section for $Ni^{58}(n,2n)Ni^{57}$ was also found[11] to be very much smaller than expected.

The two most important conclusions of this paper may be summarized as follows:

1. The conclusion of reference 5, that even after correcting for energetic differences, F_p/F_n is many times larger in 21.5-Mev proton-induced reactions than in 14-Mev neutron-induced reactions, is apparently verified and extended to the mass region 19 to 39. It should be noted, however, that this verification is based wholly on cases where proton emission is strongly favored energetically. Such was not the case in reference 5.

2. $(p,2p)$ cross sections are apparently very much larger than might have been expected from elementary barrier penetration considerations. For some of the heavier nuclei (mass 59–69) they are about 200–300 mb, for all of the lighter nuclei (mass 19–52) they are at least that large and in some cases much larger, for Fe^{54} the cross section is probably well over 500 mb, and for Ni^{58} it has been measured to be 680 mb. In general, these large cross sections are explained by more or less conventional methods if conclusion 1 above and Gugelot's measurements[3] of energy distributions of inelastically scattered protons are assumed; but the two most extreme cases, Fe^{54} and Ni^{58} are still very difficult to explain.

ACKNOWLEDGMENTS

The authors would like to acknowledge some very helpful discussions with J. D. Gow (University of California Radiation Laboratory) and J. A. Martin, and the continuous encouragement of R. S. Livingston.

[11] E. B. Paul and R. L. Clarke, Can. J. Phys. **31**, 267 (1953).

[12] B. L. Cohen, Phys. Rev. **98**, 49 (1955).

[13] The large $(p,2p)$ cross section in Ni^{58} was previously found by Martin, Green, and Lyon (private communication).