(p,pn) and ($p,\alpha n$) Excitation Functions

B. L. COHEN, E. NEWMAN, R. A. CHARPIE, AND T. H. HANDLEY
Oak Ridge National Laboratory, Oak Ridge, Tennessee
(Received October 8, 1953)

Excitation functions for (p,pn) and ($p,\alpha n$) reactions were measured with the internal, 23.5-Mev proton beam of the ORNL 86-inch cyclotron. Methods of obtaining homogeneous and known incident energy are described. The results indicate that, as found previously, emission of charged particles is much more probable than predicted by the statistical theory of nuclear reactions but that this cannot be explained, as has been proposed, by the lowering of the Coulomb barrier due to oscillations of the compound nucleus or by difficulties with the energy level density formula at low excitation energy.

INTRODUCTION AND THEORY

It has been recognized for some time[1–4] that emission of charged particles in nuclear reactions is generally much more probable than predicted by the statistical theory of nuclear reactions.[5] It has been variously proposed that this is due (1) to difficulties with the level density formula at low excitation energy,[6] (2) to competition of a direct interaction with compound nucleus formation,[7,8] (3) to a breakdown in a fundamental assumption of the theory,[9] and (4) to a lowering of the Coulomb barriers because of oscillations of the compound nucleus. The fourth proposal has grown out of the recent calculations of Wheeler and Hill[10,11] which indicate that surface oscillations of the compound nucleus are promoted in magnitude and controlled in direction by the effect of the incident particle. Their theory, known as the "collective model," forms a very satisfying union between the "individual particle" and "liquid drop" models, and has received experimental support from angular distribution measurements.[12,13,13a] In accordance with this model, portions of the nuclear surface spend a considerable time further than their average distance from the center of the nucleus so that charged particles can be emitted without experiencing as large a Coulomb barrier as if they were emitted from an undistorted spherical nucleus. This effect would be observable as excessively large total cross sections for nuclear reactions in which charged particles are emitted, whence proposal (4) above; and as a distortion in the energy spectrum of the emitted particles in that a larger number would have energies considerably lower than the Coulomb barrier calculated for the undistorted nucleus. The experiments described in this paper were designed specifically to determine whether this second effect is observed, and thus to decide whether proposal (4) above is valid; at the same time, however, they produce new evidence for the phenomenon of excessive emission of charged particles from nuclear reactions by observing it for new types of reactions, and in a new energy region where proposal (1) above is not applicable.

It was first pointed out by Weisskopf and Ewing[14] that excitation functions for reactions of the type (x,yn) (where x and y are any nuclear particles) can be analyzed to give the approximate energy distribution with which the particle y is emitted. This is possible because, to a good approximation, the probability for neutron emission to follow emission of y is just the probability that y is emitted with energy sufficiently low to leave neutron emission energetically possible. Thus, the cross section for an (x,yn) reaction for incident energy E is

$$\sigma(x,yn)=\sigma_x\frac{1}{\sum_i f_i}\int_0^{E-B} N_y(\epsilon)d\epsilon, \quad (1)$$

where σ_x is the cross section for capture of x to form the compound nucleus, B is the threshold for the (x,yn) reaction, $N_y(\epsilon)$ is the energy distribution with which y is emitted, $f_i=\int_0^\infty N_i(\epsilon)d\epsilon$, the total probability for the compound nucleus to decay with emission of particle i, and the sum is over all particles that can be emitted. The behavior near the threshold can best be seen by differentiating (1) with respect to E, giving

$$N_y(E-B)=\frac{d\sigma(x,yn)}{dE}\text{ plus terms in }\frac{d\sigma_x}{dE},\frac{df_i}{dE},\frac{dN_y}{dE}. \quad (2)$$

For high-energy incident particles, $d\sigma_x/dE$ is small and relatively well known; the df_i/dE and dN_y/dE terms tend to compensate, they are much smaller than the leading term, and in any case, they can be approximately corrected for. Thus, a measurement of $\sigma(x,yn)$ as a function of E gives relatively direct information on

[1] H. Waffler, Helv. Phys. Acta. **23**, 239 (1950).
[2] B. L. Cohen, Phys. Rev. **81**, 184 (1951).
[3] E. B. Paul and R. L. Clarke, Can. J. Phys. **31**, 267 (1953).
[4] S. N. Ghoshal, Phys. Rev. **80**, 939 (1950).
[5] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).
[6] B. T. Feld *et al.*, U. S. Atomic Energy Commission Report NYO-636.
[7] H. McManus and W. T. Sharp, Phys. Rev. **87**, 188 (1952).
[8] Austern, Butler, McManus, and Sharp, Phys. Rev. **91**, 453 (1953).
[9] B. L. Cohen, Phys. Rev. **92**, 1245 (1953).
[10] D. L. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953).
[11] J. A. Wheeler, invited paper at Rochester Meeting of Amercan Physical Society, June, 1953 [Phys. Rev. **92**, 843 (1953)].
[12] I. Halperin *et al.*, U. S. Atomic Energy Commission AECU-2494 (unpublished).
[13] W. L. Dickinson and J. E. Brolley, Phys. Rev. **90**, 388 (1953).
[13a] Cohen, Jones, McCormick, and Ferrell, Phys. Rev. **99**, 625 (1954).
[14] V. F. Weisskopf and D. H. Ewing, Phys. Rev. **57**, 472 (1940).

N_y. It is customary to assume N_y to be given by

$$N_y(\epsilon)=\sigma_y(\epsilon)\exp(-\epsilon/T), \quad (3)$$

where σ_y is the cross section for capture of y and T is the nuclear temperature. When y is a neutron, σ_y is approximately independent of energy so that $(x,2n)$ excitation functions give relatively sensitive determinations of T. Several measurements of this type have been reported[15–18]. In cases where y is a charged particle, σ_y is essentially the Coulomb barrier penetration factor which is strongly energy dependent, so that the measurements give good determinations of σ_y, and only rough determinations of T, being much more sensitive to the former. Thus, we see that measurements of (p,pn) and $(p,\alpha n)$ excitation functions give good determinations of the Coulomb barriers encountered by the emitted protons and alphas, and therefore provide a suitable test for the effect of the Wheeler-Hill collective model on these Colomb barriers.

If the (x,yp) threshold is considerably lower than the (x,yn), the above considerations are generally applicable to that reaction; however, the situation would be somewhat modified by the competition of γ emission with the emission of very low-energy protons.

For energies well above the threshold, it can be seen from (1) and the definition of f_i, that

$$\sigma(x,yn)=\sigma_x f_y/\textstyle\sum_i f_i, \quad (4)$$

which, according to the usual definition, is $\sigma(x,y)$. Thus, the absolute cross sections at high energies can be compared with the predictions of the statistical theory of nuclear reactions in the same way as the data from references 1–3.

EXPERIMENTAL METHODS

Although measurements of excitation functions by stacked-foil techniques are among the simplest and oldest nuclear physics experiments, the extension of the method to use with an internal, circulating cyclotron beam is not simple. Two of the most difficult problems are (a) arranging the foil stack so that all incident particles which pass through any foil must pass through all foils, and (b) obtaining monoenergetic incident particles of a known energy. These requirements were met in the following manner:

Requirement (a) essentially demands that the foil stack be placed behind a thick window frame. Since the radial distance between successive orbits in a cyclotron is quite small, it is difficult to prevent almost the entire beam from striking the window frame, and in addition, since the radial width is larger for lower energies, the small fraction of the beam that does get through is likely to contain a high proportion of the low-energy components of the circulating beam. This problem was studied in detail,[19] and it was found that by detuning the magnetic field approximately 0.2 percent, the phase of the ions circulating in the cyclotron can be shifted relative to the dee voltage sufficiently to increase the radial width of the beam to about $\frac{1}{4}$ inch. A well-grounded ion source was also found to be important.

Requirement (b) was finally met by installing a thick probe in the cyclotron 180° away from the target. This limits the maximum energy of the protons entering the window. It was found that the low-energy portion of the cyclotron beam could be reduced by operating at high dee voltage. By suitably adjusting the target position and the dee voltage, the point was finally reached where the energy distribution of the protons striking the foil had a full width at half-maximum of less than $\frac{1}{2}$ Mev, and a sharp high-energy cutoff. Such a spectrum is satisfactory for measuring excitation functions which increase with increasing energy, because in these cases, the low-energy tail is not effective.

The energy distribution of the protons entering the window was obtained by measuring the excitation function for the (p,n) reaction in copper and comparing with the accurately known excitation function[20] in the region near the threshold. The maximum energy was calculated from the magnetic field and the distance between the 180° probe and the target and was found to agree with the copper excitation function measurement if the usual correction[21] of a little over 1 percent is applied to the range-energy curves.[22]

The proton current was determined by measuring the 38-minute activity induced in copper by the (p,n) reaction. The cross section for this reaction has a broad maximum of about 530 mb near 13 Mev which is ideal for calibration purposes. Absolute beta counting was carried out with thin foils mounted on cellophane about $\frac{1}{8}$ in. below a thin-window Geiger counter. Small corrections were made for beta absorption by the methods of reference 2. As a check, a few absolute cross-section determinations were made by counting the 511-kev annihilation radiation with a scintillation spectrometer and comparing with the annihilation radiation from the 38-minute copper. The decay scheme corrections are available in the literature[23] for every case except the electron capture branching ratio for Ag^{106} which was estimated theoretically. Considering these factors and the uncertainty in the copper cross section, the probable errors of the absolute calibration are about 25 percent. Samples not being used for absolute cross-section calibration were counted under convenient absorbers (to eliminate background activi-

[15] Brolley, Fowler, and Schlacks, Phys. Rev. **88**, 618 (1953).
[16] H. C. Martin and B. C. Divey, Phys. Rev. **86**, 565 (1952).
[17] D. J. Tendam and H. L. Bradt, Phys. Rev. **72**, 1118 (1947).
[18] Bleuler, Stebbins, and Tendam, Phys. Rev. **90**, 460 (1953).

[19] B. L. Cohen, Oak Ridge National Laboratory Report ORNL-1348 (unpublished).
[20] Blaser, Boehm, Marmier, and Peaslee, Helv. Phys. Acta. **24**, 3 (1951).
[21] H. Bechsel and R. F. Mozley, Phys. Rev. **90**, 354 (1953).
[22] Aron, Hoffman, and Williams, University of California Radiation Laboratory Report UCRL-121 (Rev.) (unpublished).
[23] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

ties) or, in the case of Cu^{60}, on a scintillation spectrometer set to count γ rays above 3 Mev.

The target is made of 2-S aluminum to avoid irradiation of personnel from long-lived impurity activities. The demountable window frames and backings are of tantalum, with the former bent in an L shape to aid in foil stacking.

RESULTS AND DISCUSSION

The excitation functions for (p,pn) reactions on Cu^{65}, Pd^{110}, Ag^{107}, and Ta^{181}, and for the $(p,\alpha n)$ reaction on Zn^{64} are shown in Figs. 1 to 5. Individual runs were normalized in the 18- to 22-Mev region since absolute calibrations were made in separate runs. The dashed lines represent calculations of the relative excitation functions from Eq. (1) using $N_y(\epsilon)$ from Eq. (3) with various values of the nuclear temperature T. The σ_y in Eq. (3), which are essentially the Coulomb barrier penetration factors for the outgoing protons or alphas, were taken from reference 5, p. 352 ff. They are calculated for spherical nuclei of radius equal to $1.4A^{\frac{1}{3}} \times 10^{-13}$ cm. The dot-dash lines represent the result that would be obtained for $T=1$ Mev if the corrections discussed following Eq. (2) were not taken into account. The magnitude of these corrections—which is the most uncertain factor in the analysis of the experiments—can be seen by comparing these curves with the dashed curves labeled $T=1$ Mev.

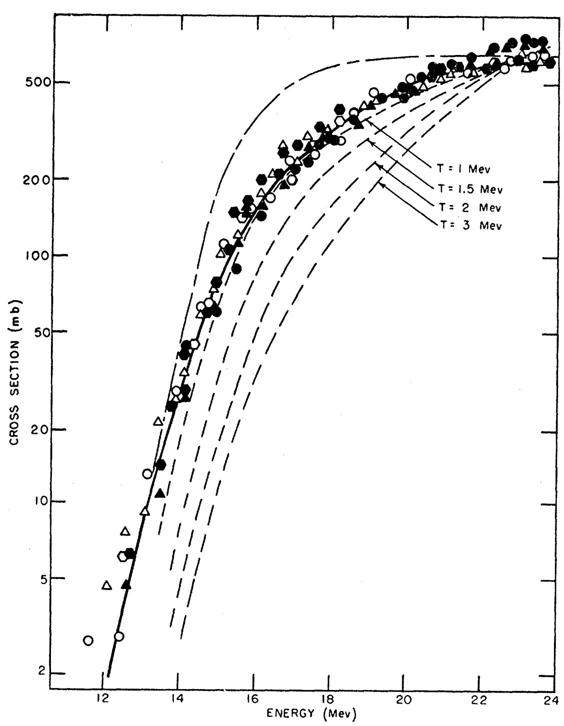

FIG. 1. Excitation function for $Cu^{65}(p,pn)Cu^{64}$. The data also include a small contribution from the (p,d) reaction as discussed in text. Dashed lines are theoretical predictions of statistical theory for various assumed nuclear temperatures, all normalized at 23 Mev. The dot-dash line is the curve for $T=1$ Mev obtained by neglecting all but first term in Eq. (2). Calculations are for nuclear radii of $1.4A^{\frac{1}{3}}\times10^{-13}$ cm. Data from various runs are normalized to superpose between 18 and 22 Mev. Absolute cross sections have probable errors of about 25 percent. Data are corrected for $(n,2n)$ reactions due to neutron background.

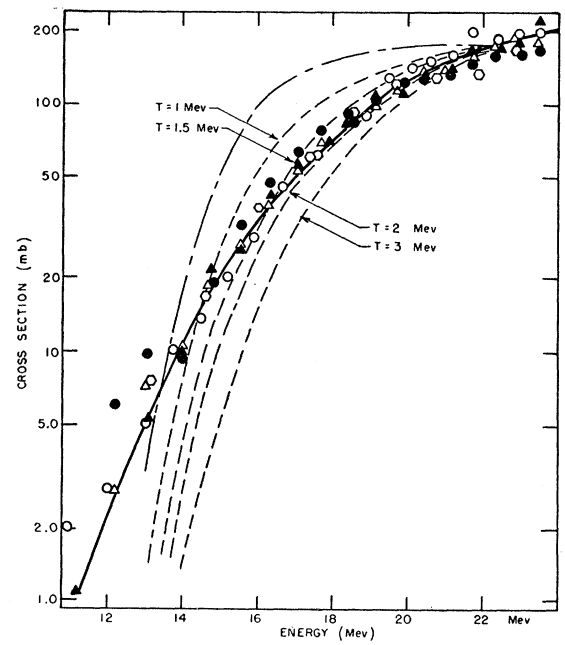

FIG. 2. Excitation function for $Pd^{110}(p,pn)Pd^{109}$. See caption for Fig. 1.

Figure 6 shows the excitation function for the (p,α) reaction on Zn^{64}. This excitation function decreases with increasing proton energy over a large region, so that the low-energy tail in the incident energy distribution distorts the data by a fraction of a Mev, but they can be considered at least semiquantitatively valid. The theory indicates that the dropoff in the (p,α) cross section is due to the onset of the $(p,\alpha p)$, so that the shape of the $(p,\alpha p)$ excitation function can be inferred and compared with the theory. The dashed curve in Fig. 6 labeled "$(p,\alpha p)$, $T=3$ Mev" is the theoretical $(p,\alpha p)$ excitation function calculated by the same method as that for the $(p,\alpha n)$; the fact that the experimental curve is displaced to the right of it indicates that γ emission competes favorably with emission of protons with less than about one to two Mev.

Figure 6 also shows the $(p,\alpha n)$ curve from Fig. 5. In order to calculate the $(p,\alpha n)$ threshold from mass data, it was necessary to know the threshold for the reaction $Ni^{60}(p,n)Cu^{60}$. This was measured by bombarding a stack of interleaved nickel and copper foils and comparing the two excitation functions. The $Ni^{60}(p,n)$ threshold was determined to be 6.7 ± 0.4 Mev.

Since for both the $(p,\alpha n)$ and $(p,\alpha p)$ cases, the measurements are in essential agreement with the theoretical curves or at least do not lie to the left of them, and since the theoretical curves were calculated

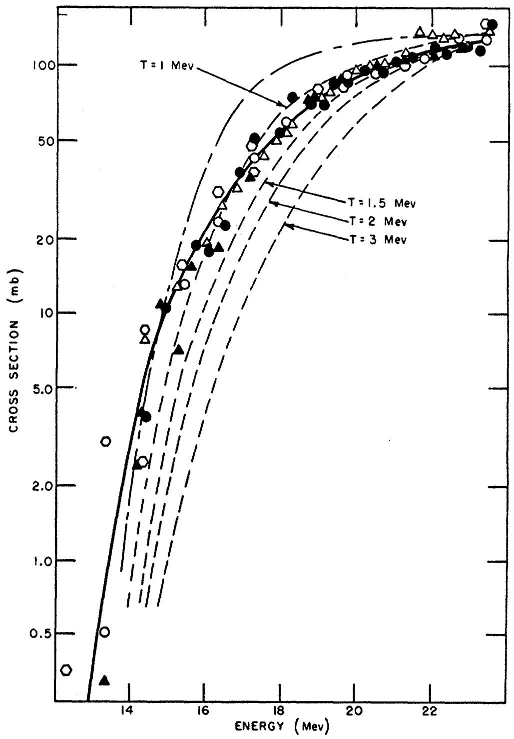

FIG. 3. Excitation function for $Ag^{107}(p,pn)Ag^{106}$. See caption for Fig. 1. Only the 24-minute isomer of Ag^{106} was observed. To correct for the other isomer, absolute values should be multiplied by about 1.6 (see reference 17). The theoretical correction for K capture has been applied.

for undistorted spherical nuclei, it is quite evident that there is no appreciable lowering of the Coulomb barriers faced by the outgoing alphas due to surface oscillations of the compound nucleus as suggested by the collective model. For example, if the Coulomb barrier were reduced by a factor of two by these oscillations as has been proposed,[11] the $(p,\alpha n)$ excitation function would rise steeply beginning at about 14 Mev, and the $(p,\alpha p)$ rise would begin between 8 and 10 Mev. Such an assumption is thus contradicted very directly by the data. This conclusion should not, of course, be construed as a contradiction of the general theory of the collective model (actually, no conclusive calculations of the expected effect have been made), but only of the proposal to extend its application to this phase of nuclear reaction theory and of the preliminary report of an experimental verification of that proposal.[11]

The absolute total (p,α) cross section at 13 Mev is in reasonable agreement with predictions of the theory. However, this is not a good case for comparison with the theory because threshold complications are extreme, the energy available for alpha emission being 12 Mev higher than for neutron emission.

The lines through the (p,pn) cross section data are shown in Fig. 7 along with the theoretical absolute values at 22 Mev. The only case where the competition between proton and neutron emission is affected by the nature of the nuclei involved is for palladium where the products of the (p,n) and (p,p) reactions are odd-odd and even-even, respectively; the theoretical (p,pn) cross section was divided by four (in Fig. 7) to correct for this.

Before drawing conclusions from Fig. 7, some account must be taken of (p,d) reactions which are known to take place by a "pick-up" process, producing the same end product as (p,pn) reactions. Attempts were made to measure absolute (p,d) cross sections by determining the outgoing deuteron flux with activation detectors.[24,25] These indicated that no more than 25 percent of the measured copper (p,pn) cross section is due to the (p,d) reaction. For the heavier elements, the method was not sufficiently sensitive.

It should be noted, however, that (p,d) reactions have thresholds 2.2 Mev lower than (p,pn) reactions, and the barrier encountered by deuterons is about 1.5 to 2.0 Mev lower than for protons.[26] Thus, if all deuterons were emitted with the maximum available energy, the excitation function would be of roughly the same shape as the theoretical (p,pn) curves but shifted 3 or 4 Mev to the left. If, on the other hand, they were emitted with a Maxwellian energy distribution, the general shape of the excitation function would be very different from the observed ones. Although the possibility cannot be

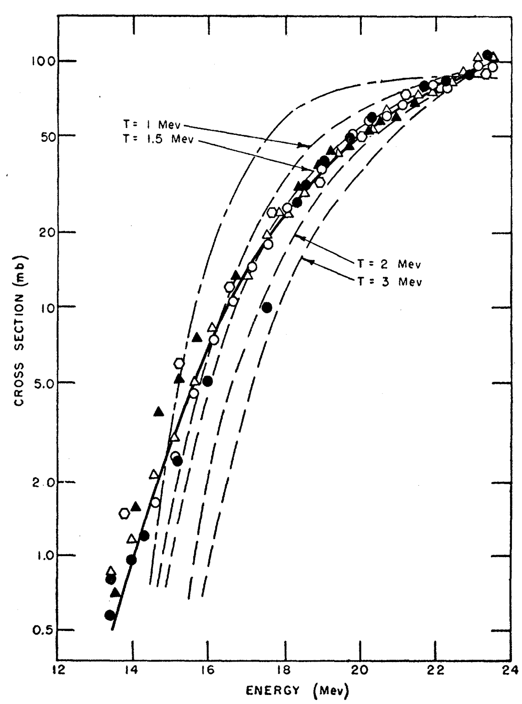

FIG. 4. Excitation function for $Ta^{181}(p,pn)Ta^{180}$. See caption for Fig. 1.

[24] B. L. Cohen and R. V. Neidigh, Rev. Sci. Instr. **25**, 255 (1954).

[25] Cohen, Newman, Handley, and Timnick, Phys. Rev. **90**, 323 (1953).

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

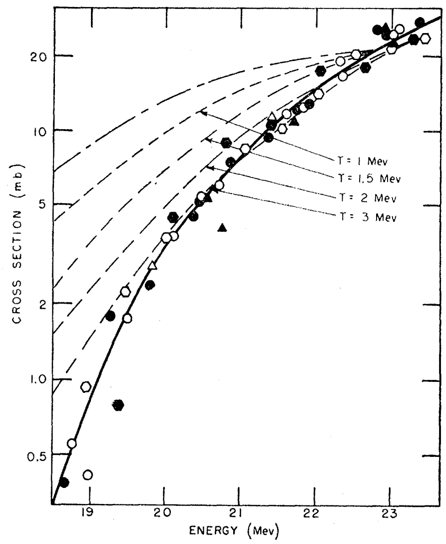

FIG. 5. Excitation function for $Zn^{64}(p,\alpha n)Cu^{60}$. See caption for Fig. 1.

logically excluded, it would require a very remarkable combination of coincidences for the spectrum of emitted deuterons to be such that the excitation functions for the (p,d) reactions reproduce so closely the theoretical excitation functions for the (p,pn) reactions.

With these facts in mind, it seems quite evident from Fig. 7 that the following conclusions can be drawn:

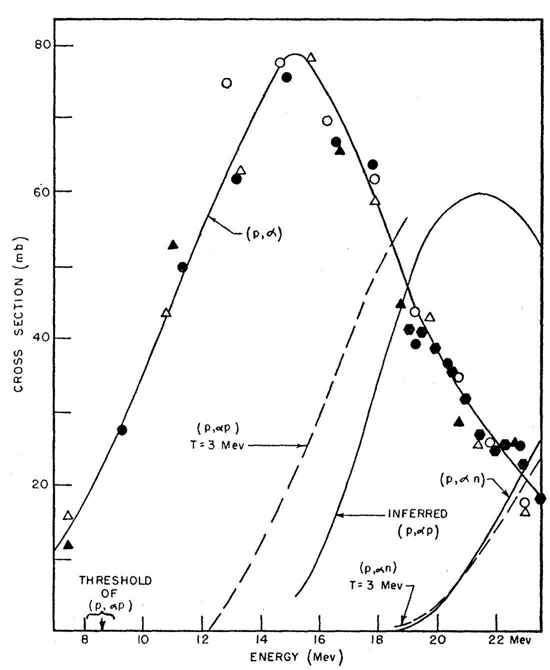

FIG. 6. Excitation functions for (p,α) and $(p,\alpha n)$ reactions on Zn^{64}. The $(p,\alpha p)$ excitation function is inferred from the dropoff in the (p,α) cross section. The $(p,\alpha n)$ curve is taken from Fig. 5. The arrow at 8.5 Mev should be captioned "Threshold for $(p,\alpha n)$."

(a) As in the case of alpha particles, the full Coulomb barrier, as calculated for spherical, undistorted nuclei, is effective in suppressing the emission of low-energy protons, contrary to expectations from the collective model. This effect is most pronounced in the case of tantalum where a reduction in the Coulomb barrier by a factor of two would essentially cause the observed excitation function to be displaced to the left of the theoretical curves by about 5 Mev, contrary to observation. It is especially worth noting that this conclusion is independent of whether or not (p,d) reactions are important contributors to the observed activities (unless, of course, they are overwhelming contributors).

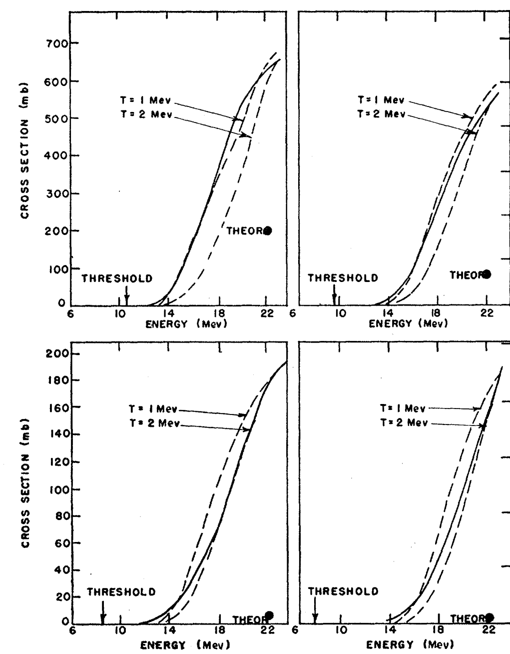

FIG. 7. (p,pn) excitation functions. The curves are lines through the data of Figs. 1–4. Solid circles are absolute cross sections at 22 Mev, calculated from the statistical theory of nuclear reactions. The upper curves are for copper (left) and silver (right); the lower curves are for palladium (left) and tantalum (right).

(b) The absolute value of the inelastic proton scattering cross sections are many times larger than the predictions of the statistical theory. This is another manifestation of the well known phenomenon of charged-particle emission being excessively probable. However, it should be noted that in previous observations of the effect,[1–3] neutron emission was energetically favored, whereas in these cases, proton emission is energetically favored. Also, the threshold complication of reference 4 is not present. The calculations are quite sensitive to these points. In addition, the calculations on these data are considerably less sensitive to the choice of nuclear temperatures. They are completely independent of the behavior of energy-level densities at low excitation energies because (p,pn) reactions are not energetically possible unless the residual nucleus (after emission of the proton) has sufficient excitation to allow neutron emission. Actually most of the reactions leave the residual nucleus with considerably higher excitation than this minimum.

Therefore, these experiments show quite conclusively that neither the first nor the fourth proposals listed in the introduction of this paper are the correct explanations for the excessive emission of charged particles in nuclear reactions. On the other hand, they provide a new and clear-cut demonstration of that effect in an energy region where uncertainties in nuclear temperatures and in the method of correcting for thresholds cannot be important factors.

The authors would like to acknowledge the help of B. L. Ferrell and E. L. Olson in various parts of the data accumulation, and the general advice and encouragement of J. L. Fowler and R. S. Livingston.

PHYSICAL REVIEW VOLUME 94, NUMBER 3 MAY 1, 1954

Angular Distributions of Fission Fragments from 22-Mev Proton-Induced Thorium Fission

B. L. COHEN, W. H. JONES,* G. H. MCCORMICK,† AND B. L. FERRELL
Oak Ridge National Laboratory, Oak Ridge, Tennessee

(Received January 11, 1954)

The angular distributions of barium, strontium, zirconium, ruthenium, and silver fission products from thorium fission induced by 22-Mev protons were measured using the internal, circulating beam of the ORNL 86-inch cyclotron. Within the accuracy of the measurements, all angular distributions are symmetric about 90° and may be well fitted to $I(\theta)=a+b\cos^2\theta$. For Ba, Sr, Zr, Ru, and Ag fission products, for which the fission mass ratios are 1.53, 1.52, 1.37, 1.19, and 1.04, the anisotropy (b/a) is 0.26, 0.25, 0.19, 0.15, and 0.10, respectively.

INTRODUCTION

THE angular distributions of fragments from fission induced by thermal neutrons should be isotropic according to the Eisner-Sachs-Yang rule,[1] and the fact that they are, has been experimentally confirmed.[2] It has commonly been assumed that angular distributions are also isotropic for fission induced by high-energy particles, because fission proceeds by a compound-nucleus interaction in which the number of intermediate and final states is sufficiently large that the effects of individual levels, which are the usual cause of anisotropic angular distributions, would be expected to average out. This expectation, however, has not been verified by experiment.

The first reported measurements of this type was the work of Winhold, Demos, and Halpern[3] on thorium photofission; their data indicate angular distributions of the form $I=\alpha+\beta\sin^2\theta$, where β/α, which is a measure of the anisotropy, reaches values as large as 1.2. They interpret the phenomenon as due to the effects of dipole absorption. However, the recent theoretical work of Wheeler and Hill[4] has shown that compound nuclei undergo large oscillations which lower the Coulomb barrier in specified directions and thereby explain, at least qualitatively, the results of reference 3. At their suggestion, Dickinson and Brolley[2] measured the 0°/90° intensity ratios of fragments from 14-Mev neutron-induced fission of Th^{232}, Np^{237}, U^{233}, U^{235}, and U^{238}. In all cases, they found these ratios to be greater than unity (1.2−1.5), which is in qualitative agreement with the Wheeler-Hill prediction for particle-induced fission.

To throw further light on this problem, a program for the measurement of angular distributions of the products of *proton*-induced fission was undertaken by utilizing the internal, circulating beam of the ORNL 86-inch cyclotron. This approach has the advantage of providing very large incident particle currents, and is especially timely because methods of measuring angular distributions with this beam have recently been studied in considerable detail.[5] In this paper, we describe measurements of angular distributions of Ba, Sr, Zr, Ru, and Ag fission products from 22-Mev proton-induced thorium fission. The work is now being extended to other target elements, and eventually will be extended to other bombarding energies.

EXPERIMENTAL PROCEDURE AND RESULTS

The target assembly is similar to that pictured in reference 5, except that a more accurate method of

* Research participant from Chemistry Department, Emory University, Atlanta, Georgia.
† Research participant from Chemistry Department, University of Denver, Denver, Colorado.

[1] E. Eisner and R. G. Sachs, Phys. Rev. **72**, 680 (1947); C. N. Yang, Phys. Rev. **74**, 764 (1948).
[2] W. L. Dickinson and J. E. Brolley, Phys. Rev. **90**, 388 (1953).
[3] Winhold, Demos, and Halpern, Phys. Rev. **87**, 1139 (1952); I. Halpern and E. J. Winhold, U. S. Atomic Energy Commission Report AECU-2494 (unpublished).
[4] D. L. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953).
[5] B. L. Cohen and R. V. Neidigh, Rev. Sci. Instr. **25**, 255 (1954).