Implications of the Photonuclear Effect in Zr^{90}†

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The reactions $Zr^{90}(\gamma,n)Zr^{89}$ and $Zr^{90}(\gamma,n)Zr^{89m}$ have been studied in detail. The threshold for $Zr^{90}(\gamma,n)Zr^{89m}$ is 12.37±0.09 Mev; that for $Zr^{90}(\gamma,n)Zr^{89}$ is 11.78±0.09 Mev. A discussion is given of the precision of (γ,n) threshold determinations and of the error sometimes introduced because of large spin differences. The detailed shape of the (γ,n) cross section is used to obtain data on both the absorption mechanism of gamma rays and the competition between the emission of neutrons and gamma rays by a compound nucleus. At about 12.4 Mev in Zr^{90}, less than 60% of the absorption occurs by the $E1$ process if $E2$ is the only other absorption process. If both $M1$ and $E2$ absorption occur equally, only about 10% of the absorption is $E1$. The competition between gamma ray and neutron emission could not be obtained quantitatively but the results are consistent with other quite different measurements of this competition.

INTRODUCTION

THIS paper presents the results of a detailed experiment on the reactions $Zr^{90}(\gamma,n)Zr^{89m}$ and $Zr^{90}(\gamma,n)Zr^{89}$ together with an interpretation of the results. The main purpose of the experiment was to determine the extent to which photonuclear reactions could be used to study specifically nuclear properties such as neutron binding energies, spins and parities, neutron and gamma-ray widths, energy level densities, etc. The quantitative comparison shown below between experiment and theory clearly demonstrates the importance of the centrifugal barrier in reducing neutron emission; this has the following six implications:

(1) The photonuclear effect can be used to obtain quantitative data about the competition between neutron and gamma-ray emission. Not only will such data supplement comparable data obtainable from the absorption and scattering of neutrons but they will also provide a test of the theory of the compound nucleus by indicating whether states activated by photon absorption have any special tendency to be de-excited by photon emission.

(2) Because the centrifugal barrier so strongly inhibits the emission of neutrons which carry away large angular momenta, quantitative photonuclear experiments provide a sensitive means for determining the fraction of photon absorption that occurs through electric quadrupole or magnetic dipole interaction.

(3) The energy dependence of the (γ,n) cross section near threshold, being governed by the orbital angular momentum carried away by the neutron, can sometimes be used to determine the relative spin and parity of the daughter nucleus.

(4) Measured threshold energies for (γ,n) reactions may sometimes be appreciably higher than the actual neutron binding energy. If the ground-state spins of the parent and daughter nuclei are sufficiently different, neutron emission will not be observable until enough energy is available to leave the (γ,n) daughter nucleus in an excited state that has a more favorable spin.

(5) The photoneutron yield, Y, near threshold is not necessarily governed by a power law of the form $Y \propto (E_\beta - E_{\text{th}})^m$, where E_β is the energy of the bremsstrahlung producing electrons, E_{th} is the threshold energy and m is a positive constant. If E_1 is chosen so that the yield is fitted to a power law of the form $Y \propto (E_\beta - E_1)^m$, the energy, E_1, can be determined rather precisely but, unfortunately, there is little theoretical justification for associating E_1 with E_{th}.

(6) The study of the relative production of two energy levels in a single daughter nucleus might give information about the energy level densities of states of different spin if more were known about gamma-ray cascading.

This paper is divided into six sections. Section I shows explicitly how the photonuclear effect is governed by the nature of the gamma-ray absorption and the properties of the nucleus. This section also lists the reasons for choosing Zr^{90} for this study. Section II describes briefly the experimental procedure. Section III presents the uninterpreted experimental results and compares them with other related experimental data. Section IV contains an interpretation of the experimentally determined energy dependence of photoneutron emission near threshold. It shows the extent to which the experimental results give information about neutron and gamma-ray widths and about the multipolarity of absorbed gamma rays. Section V extends the implications of these interpretations to the general problem of determining thresholds for (γ,n) reactions. Section VI interprets the experimental results at energies well above the threshold energy (i.e., those results not discussed in either Sec. IV or V).

I. CHOICE OF PROBLEM

The principal motivation of this experiment was a desire to use photonuclear experiments as a tool to learn about nuclear energy levels. It seemed interesting to see to what extent photonuclear experiments agreed quantitatively with those parts of the theory which have had some independent experimental confirmation.

It was decided to study the energy dependence of

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photoneutron emission just above the threshold energy since in that energy range the limited number of competing processes makes a quantitative comparison with theory possible. This quantitative comparison seems feasiable even though both the multipolarity of the absorbed gamma ray and the competition between the emission of neutrons and gamma rays are not completely specified. At higher energies, the variety of competing processes (including multistep reactions) makes theoretical comparison impossible even though the large cross section clearly implies that the absorption is predominantly electric dipole.

An analysis of the experimental results in the threshold region can be made by assuming that photoneutron emission occurs through compound nucleus formation. This assumption makes it possible to deal separately with the absorption and the emission process. The absorption involves unknown fractions of electric dipole ($E1$), magnetic dipole ($M1$), and electric quadrupole ($E2$) processes. If the parent nucleus has a spin parity of 0+, the spin-parity of the compound nuclear state is 1− for $E1$ absorption, 1+ for $M1$ absorption, and 2+ for $E2$ absorption. The decay of the compound nucleus (in the energy region immediately above neutron threshold) depends on the competition between the emission of neutrons which leave the nucleus in the ground state and of gamma rays. It is known that the gamma-ray emission rate will not vary much over this narrow energy range whereas the neutron emission rate varies in a predictable manner, which depends on the angular momentum of the emitted neutrons.[1] The angular momentum of the emitted neutron, in turn, depends on the spins and parities of both the ground state of the daughter nucleus and the excited states in the compound nucleus. In general, for each type of state reached by a particular type of multipole absorption, there will be a different competition between neutrons and gamma rays predicted by theory. Thus, a study of the actual yield of photoneutrons should lead to information about both the absorption mechanism and the competition in decay.

The results obtained in this photonuclear study on zirconium near threshold do not, in themselves, determine uniquely both the multipolarity of the absorbed gamma ray and the competition between modes of decay. Instead, the results can be interpreted either to give the multipolarity if the competition is known or to give the competition if the multipolarity is known. For example, if the values of the partial widths for gamma-ray emission and neutron emission are taken from neutron absorption studies, the zirconium experimental results can be used to determine the percentage of electric dipole absorption.

[1] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952), p. 358 ff.

Choice of Isotope

The arguments presented in the foregoing make it clear that one should study the photoneutron emission cross section in an energy region in which neutron emission is competing with gamma-ray emission. If neutron emission were much more probable than gamma-ray emission, a neutron would be emitted each time a nucleus was excited and the energy dependence of photoneutron emission would merely indicate the energy dependence of the excitation of the nucleus (i.e., the energy dependence of the absorption of a gamma ray). One can insure a relatively large energy range in which neutron emission does not overwhelm gamma-ray emission by choosing an isotope with a large spin difference between the ground states of the parent and daughter nuclei so that neutron emission would be inhibited.

Since a large centrifugal barrier which inhibits neutron emission also makes it difficult to measure the (γ,n) threshold energy directly, an independent, indirect measurement of the (γ,n) threshold is required. A convenient determination of the (γ,n) threshold of the ground state is possible if the daughter nucleus also has an isomeric level which can be reached easily by neutrons. The (γ,n) threshold of the isomer can then be measured (with the same betatron to avoid calibration errors) and the (γ,n) threshold of the ground state can be inferred using the known radioactive decay of the upper isomeric level. Additional desirable characteristics of a suitable daughter nucleus for this type of experiment include conveniently measurable radioactivities and a large energy separation between isomeric levels.

Zr^{90} was the most satisfactory isotope that could be used as a target (parent) nucleus. Since Zr^{90} is an even-even nucleus, its spin parity is 0+. The radioactivities of the daughter isomers, 79-hr Zr^{89} and 4.4-min Zr^{89m} can be measured conveniently and differentiated from radioactivities produced by gamma rays absorbed by the other isotopes of zirconium. The 79-hour ground state of Zr^{89} has a spin parity of 9/2+, arising from a $(g_{9/2})^{-1}$ neutron configuration. The isomeric level, 4.4-minute Zr^{89m}, is 588 kev above the ground state and is a $\frac{1}{2}-$ level arising from a $p_{1/2}$ neutron configuration.

II. EXPERIMENTAL PROCEDURE

The experiment consisted of irradiating zirconium with bremsstrahlung x-rays from a betatron; the energy of the electrons in the betatron was varied from 11.8 Mev to 22.5 Mev. At each energy, the resultant radioactivity was measured for each of the two isomeric levels of Zr^{89}.

(A) Samples

The samples were made of natural zirconium in the form of either metal foils 1 inch square and 0.52 g/cm^2, or pressed disks of ZrO_2 powder, 1.5 inches in diameter

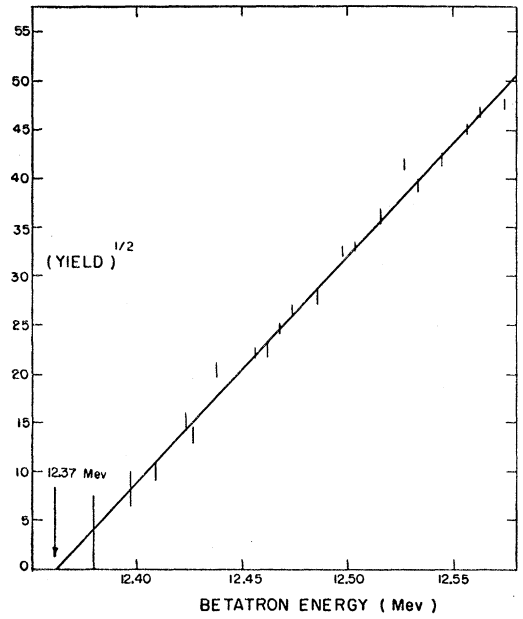

FIG. 1. $Zr^{90}(\gamma,n)Zr^{89m}$ threshold determination.

and 1.50 g/cm^2. The pressed disks were coated with an acrylic spray to prevent flaking. For the set of measurements that was made within 600 kev of the ground-state threshold energy, each sample consisted of five disks and was therefore 7.5 g/cm^2.

The samples were irradiated in a cadmium-shielded holder positioned just outside of the betatron doughnut at a distance of 22 centimeters from the betatron target. During measurements of the 4.4-minute activity, the samples were usually irradiated for five minutes. The irradiation time required for measurement of the 79-hr activity varied from 30 minutes at the highest energy to about 8 hours at energies just above 12.4 Mev. In addition, several 40-hour irradiations were made between 11.8 and 12.4 Mev to detect the very weak ground state activities that were formed below the threshold for the excitation of the isomeric level.

(B) Beam Monitors

Two different monitoring procedures were used during the course of these experiments. The earliest measurements were made using a Victoreen ionization thimble [calibrated to measure roentgens (r) of radiation] in the center of an 8-cm Lucite cube. The Victoreen thimble was placed either 1 meter or 3 meters from the betatron target. The average intensity of the betatron beam at an energy of 22 Mev as measured by this monitor at one meter was 75 r/minute.

The more recent measurements were made using an aluminum-walled air-filled ionization chamber which was placed behind 2 inches of lead and was positioned 59 cm from the betatron target so that both the sample and the ionization chamber subtended the same portions of the x-ray beam. The ionization current was measured with the aid of a vibrating reed electrometer circuit.[2] The x-ray response of the ionization chamber as used (i.e., behind 2 in. of Pb) was found to be the same as the response of the Victoreen thimble to within 1%.[3] For the 4.4-minute measurements, the electrometer circuit contained a time constant equal to the mean life of the radioactivity so that fluctuations in the beam intensity during a bombardment did not introduce any error. All of the results quoted in this paper were obtained with the ionization chamber monitor. However, the Victoreen thimble was used in measurements of the threshold energies and of the yields above threshold. These measurements, although less precise than the final results, were consistent with them.

(C) Detectors and Counting Equipment

Although both Geiger Counters and NaI(Tl) scintillation detectors were used to measure the radioactivities, the main data were obtained with the scintillation detectors. Measurements made with Geiger counters were considerably less precise than the scintillation measurements; the Geiger counter data were therefore used merely as an indication that no gross errors or backgrounds were introduced by scintillation detectors.

The detection of gamma rays by scintillation counters makes possible the effective use of thick samples; this is feasible because gamma-ray emission is the predominant mode of radioactive decays. For the 79-hour Zr^{89}, both the 75% K-capture branch and the 25% positron branch lead to a 14-second, 913-kev isomeric level in Y^{89} which decays by gamma-ray emission.[4] The positrons were all stopped either in the source or in surrounding Bakelite so that the 511-kev annihilation quanta could be detected by the NaI crystal. In the case of 4.4-min Zr^{89m}, 86% of the decays emit 588-kev gamma rays, 7% emit conversion electrons and the remaining 7% are K-capture or positron events which proceed through a 1.53-Mev gamma-ray emitting level in Y^{89}.[5] Gamma-ray detection was not only more efficient but was also advantageous in minimizing the background produced by photoinduced activities in other isotopes of zirconium.

A single-channel pulse-height analyzer was used to reduce further the effect of background and of the other photo-induced activities. For the 4.4 min, Zr^{89m} detection, only pulses corresponding to the 588-kev photopeak were counted. The 79-hr Zr^{89} activity was measured by counting either the 913-kev gamma ray's photopeak pulses or by counting all the pulses in and between the 511-kev and the 913-kev photopeaks. The most precise measurements were made by obtaining the pulse-height distribution in the region of the 913-

[2] Vibrating Reed Electrometer, Model 30, Applied Physics Corporation, Pasadena, California.
[3] A. S. Penfold, Ph.D. thesis, University of Illinois, 1955 (unpublished).
[4] M. Goldhaber *et al.*, Phys. Rev. **83**, 661 (1951).
[5] F. J. Shore *et al.*, Phys. Rev. **91**, 1203 (1953).

kev photopeak. This pulse-height distribution was needed in order to subtract properly the effect of the 721-kev[6–8] and 754-kev[7,8] gamma rays emitted by 65-day Zr^{95} and the 764-kev gamma ray[7,8] emitted by its 35-day Nb^{95} daughter.

In order to determine the amount of 79-hour activity and separate it from the longer lived background, it was necessary to count each sample from time to time over a period of several weeks. Two completely independent sets of counting equipment were maintained in order to guard against equipment failure over these long periods. Each set consisted of a NaI(Tl) scintillation crystal, a Dumont 6292 photomultiplier, a stabilized high-voltage power supply,[9] a linear amplifier and preamplifier, a single-channel pulse-height selector,[9,10] a scaler, and a Streeter-Amet automatic count recorder.

Although the equipments were reliable and relatively stable, there was a slow drift in the voltage amplitude of the output pulses from the amplifier. This drift was not traced in detail but was probably due to temperature fluctuations which affected both the high-voltage supply and the amplifier. The effects of drifts were minimized by positioning the pulse-height selector so that the counting rate was insensitive to small changes in pulse amplitude. In addition, the high voltage was readjusted (usually by less than 0.5% per day) if the over-all gain shifted by as much as 1%.

Three different NaI crystals were used during the entire set of experiments. Two crystals were cylindrical; one was $\frac{3}{4}$ in. in diameter and 1 in. long while the other was 1 in. in diameter and 1.5 in. long. The third crystal, which was most efficient and was used in the most precise measurements, was a rectangular prism, 1 in. $\times$1 in.$\times$2 in. Each crystal was mounted on optically flat glass with a bonding agent[11] and sealed in a thin-walled aluminum container. A layer of MgO between the crystal and the aluminum container served as a diffuse reflector. The crystals all had a resolution of about 10% for the Cs^{137} 662-kev gamma ray; the largest crystal had a photopeak-to-valley counting rate ratio of 25. The crystals and phototubes were enclosed in a Pb house which had walls and ceilings 4 in. thick to reduce the cosmic-ray background.

III. EXPERIMENTAL RESULTS

It is convenient to divide the experimental results into four categories: (A) standard threshold measurement of the 4.4-minute activity, (B) standard threshold measurement of the 79-hour activity, (C) careful determination of the yield of $Zr^{90}(\gamma,n)Zr^{89}$ in the energy region within 575 kev of threshold, and (D) standard yield measurements from 12.5 Mev to 22.5 Mev.

(A) 4.4-Minute Threshold Measurement

The threshold of $Zr(\gamma,n)Zr^{89m}$ was measured by studying the yield of 4.4-min Zr^{89m} as a function of energy. The threshold was determined relatively precisely on seven different occasions during a two year period. Each determination consisted of finding the yield at a set of different energies spaced about 18 kev apart as shown by the set of data in Fig. 1. In addition, both the $Cu^{63}(\gamma,n)Cu^{62}$ threshold and an $O^{16}(\gamma,n)O^{15}$ check point were taken on the same day as each of the threshold determinations.

In addition to these seven independent threshold measurements, there were ten other precise measurements of the energy dependence of the $Zr^{90}(\gamma,n)Zr^{89m}$ yield in the threshold region. These ten auxiliary measurements are not considered satisfactory threshold determinations because they were not accompanied by complete calibrations of the betatron. These measurements were never inconsistent with the threshold determinations and were used to check on the stability of the betatron rather than to determine the threshold. These measurements also confirm the dependence of the yield on the square of the energy above threshold [i.e., yield $(E_\beta) \propto (E_\beta - E_{\text{th}})^2$, where E_β = betatron energy and E_{th} = threshold energy].

The "integrator setting" of the $Zr^{90}(\gamma,n)Zr^{89m}$ threshold was 2044$\pm$3 helipot units (each unit corresponds to about 6.15 kev). Part of this 3 "unit" error comes from the uncertainty of the proper extrapolation procedure as will be explained in Sec. V.

On this helipot unit scale, the $Cu^{63}(\gamma,n)Cu^{62}$ threshold is 1773$\pm$2 units, the calibrating $O^{16}(\gamma,n)O^{15}$ "break" is 2800$\pm$2 units, and the $O^{16}(\gamma,n)O^{15}$ threshold is 2567$\pm$3 units.[12] The $Cu^{63}(\gamma,n)Cu^{62}$ threshold has been determined independently by M. Birnbaum[13] and by the Saskatoon (Saskatchewan) group[14] as 10.61$\pm$0.05 Mev and 10.73$\pm$0.05 Mev, respectively. Following Penfold and Spicer,[12] we use the value 10.73$\pm$0.05 Mev. The $O^{16}(\gamma,n)O^{15}$ threshold is known to be 15.605 $\pm$0.012 Mev.[15] If it is assumed that the betatron energy scale is linear, the difference between the $Zr^{90}(\gamma,n)Zr^{89m}$ threshold and the $Cu^{63}(\gamma,n)Cu^{62}$ threshold is 1.66$\pm$0.04

[6] H. Slatis and L. Zappa, Arkiv Fysik **6**, 81 (1953).
[7] J. M. Cork *et al.*, Phys. Rev. **90**, 579 (1953).
[8] P. S. Mittelman, Phys. Rev. **94**, 99 (1954).
[9] We are indebted to W. A. Higinbotham and R. L. Chase, both of Brookhaven National Laboratory, for the design of, and many useful discussions about, the high-voltage supply and the pulse-height selector.
[10] A pulse-height selector extremely similar to the one designed by R. L. Chase and used in this experiment is built by the Atomic Instrument Company of Cambridge, Massachusetts.
[11] Bonding Agent *R*-313, C. H. Biggs and Company, 11616 West Pico Boulevard, W. Los Angeles, California.
[12] The position of the $O^{16}(\gamma,n)O^{15}$ threshold was measured by A. S. Penfold and B. M. Spicer who have done very careful work on the energy calibration of the University of Illinois Betatron. The calibration points which we used were the more precisely determinable ones at 1773 and 2800 integrator or helipot units. We base our energy determination on these secondary standards which are justified in: A. S. Penfold Ph.D. thesis, University of Illinois, 1955 (unpublished) and B. M. Spicer, Ph.D. thesis, University of Melbourne Australia, 1955 (unpublished).
[13] M. Birnbaum, Phys. Rev. **93**, 146 (1954).
[14] Robinson, McPherson, Greenberg, Katz, and Haslam (to be published).
[15] F. Ajzenberg and T. Lauritsen, Revs. Modern Phys. **24**, 321 (1952).

Mev. Using the Cu threshold as 10.73±0.05 Mev, we get a value of 12.39±0.075 Mev for the threshold of $Zr^{90}(\gamma,n)Zr^{89m}$, where the error given represents the extreme limit. (If the Cu threshold were known as 10.73±0.02 Mev, the Zr-Cu difference would be 1.66 ±0.03 and the Zr threshold energy would be 12.39 ±0.055.) An additional complication in the determination of the threshold arises because Penfold and Spicer[12] claim that the Illinois betatron calibration is not linear and they use a cubic calibration function for the betatron. Using their calibration function we would get, as a threshold energy, 12.36 Mev with an error of about ±65 kev. Until the details of the betatron calibration are understood better, the $Zr^{90}(\gamma,n)Zr^{89m}$ threshold can be conservatively stated as 12.37±0.09 Mev. It should be noted that the quoted error is $4\frac{1}{2}$ times the uncertainty in the determination and is therefore mostly due to the unsolved difficulties of calibrating a betatron precisely.

The threshold value of 12.37±0.09 Mev is in good agreement with the value of 12.48±0.15 quoted by an earlier Illinois group,[16] particularly since the earilier work used 10.9 Mev as the Cu^{63} threshold. The present measurement is also in good agreement with the value of 12.27±0.09 calculated from the $Y^{89}(d,p)Y^{90}$ Q-value of 4.41 Mev[17] and the best values of the beta decays of Y^{90} and Zr^{89}.[5,18] However, the results are somewhat outside the quoted errors of the values of 12.81±0.35 Mev (arising from the measured masses[19] of Zr^{90} and Y^{89}) and 12.1±0.1 Mev (which comes from a betatron threshold determination[20]).

Note added in proof.—Our $Zr^{89}(\gamma,n)Zr^{89m}$ threshold is also in agreement with the measurement of the Sao Paulo, Brazil group which was recently reported at the International Conference on the Peaceful Uses of Atomic Energy (U.N. 897). Their value of 12.20±0.06 Mev differs from ours only because their measured $Cu^{63}(\gamma,n)Cu^{62}$ threshold is 10.54 Mev instead of 10.73 Mev which we have adopted.

(B) 79-Hour Threshold Measurement

Once the 4.4-minute threshold is known to be 12.37 ±0.09 Mev, the threshold for the 79-hour ground-state reaction, $Zr^{90}(\gamma,n)Zr^{89}$, is known from radioactivity[5] to be 11.78±0.09 Mev. This value agrees well with the value of 11.9±0.3 Mev obtained using a betatron,[20] but we shall show below that this agreement is probably fortuitous.

We proceeded to try to measure the threshold for the 79-hour activity as though its relationship to the 4.4-minute threshold were unknown. The results are shown in Fig. 2, which appears to indicate that the 79-hour threshold is equal to the 4.4-minute threshold; this determination has an apparent experimental error of ±100 kev. This error, which is quite large considering the fact that it does not include any of the errors due to uncertainties in the calibration, is mostly due to the difficulty in measuring a 79-hour activity accurately near threshold. The experimental points were taken every 300 kev for the first million electron volts above threshold and every 600 kev thereafter. Each experimental point included about 8 hours of betatron bombardment of a 1.59-g/cm², 1-in. diameter disk and several weeks of counting all the pulses (without taking a spectrum) in a scintillation spectrometer. This procedure represents more than normal care in determining a threshold.

The dotted points below the apparent threshold show the precision which could have been obtained if experimental points had been taken using the same procedure at lower energies. The three points shown below the apparent threshold with the very small statistical errors are the actual points observed using a much more precise detecting technique. These precise points were not considered in drawing the line to find the apparent threshold.

Figure 2 shows clearly that a "standard" threshold determination of the 79-hour activity would have given the threshold of the first excited state in Zr^{89} rather than the ground-state threshold.

(C) Precise Determination of the 79-Hour Yield at Low Energies

A more precise technique was used to find the actual yield of the 79-hour activity below the threshold of the 4.4-minute activity. Thick (7.5-g/cm²) $1\frac{1}{2}$-in. disks of ZrO_2 were bombarded for about 40 hours each at energies 375 kev, 475 kev, and 575 kev above the actual threshold of the $Zr^{90}(\gamma,n)Zr^{89}$ reaction. (These points were respectively 213 kev, 113 kev, and 13 kev below

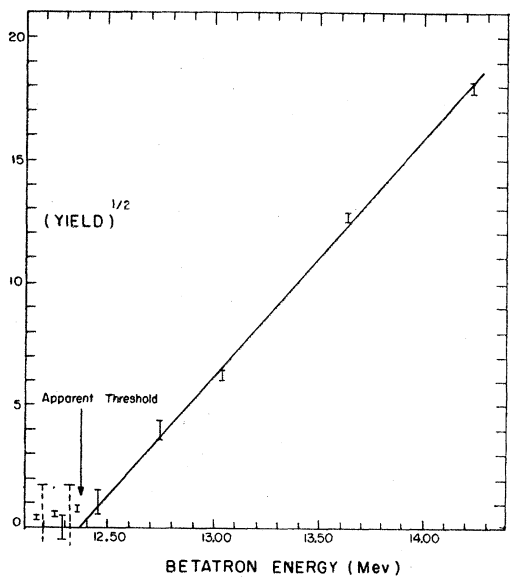

FIG. 2. $Zr^{90}(\gamma,n)Zr^{89}$ threshold determination.

[16] Hanson, Duffield, Knight, Diven, and Palevsky, Phys. Rev. **76**, 578 (1949).
[17] N. S. Wall, Phys. Rev. **96**, 664 (1954).
[18] R. W. King, Revs. Modern Phys. **26**, 327 (1954).
[19] Collins, Johnson, and Nier, Phys. Rev. **94**, 398 (1954).
[20] Ogle, Brown, and Carson, Phys. Rev. **78**, 63 (1950).

the threshold for forming 4.4-minute Zr^{89m}. The radioactivity in these three samples was then counted intermittently for three weeks using a single-channel pulse-height selector. Each pulse-height distribution curve was then analyzed in order to determine that part of the counting rate due to the gamma rays of about 750 kev from 65-day Zr^{95} and its 35-day Nb^{95} daughter.[6–8] A typical pulse-height curve is shown in Fig. 3. In order to minimize confusion, the pulse-height distributions of only three different days are shown. The actual intensity of the 79-hour, 913-kev photopeak was determined by plotting the maximum number of counts in the photopeak as a function of time and fitting these points with a 79-hour half-life curve.

For comparison, an experimental point was also taken 1163 kev above the 79-hour threshold (i.e., 575 kev above the 4.4-minute threshold). The ratio of the 913-kev activity (per roentgen) for the four points, 375, 475, 575, and 1163 kev above the 79-hour threshold, was $(0.94\pm0.20):(1.94\pm0.25):(4\pm0.4):100$. These data will be discussed and interpreted in Sec. IV.

(D) Determination of the Yield from 12.5 Mev to 22.5 Mev

The yield of the 4.4-minute activity as a function of betatron energy was quite easy to obtain. The two most precise measurements were made using metallic zirconium foils and detecting the gamma rays in the 588-kev photopeak. These measurements, which agreed within statistics, were combined in order to extract a cross section as shown in Sec. VI. One of these 4.4-minute activation curves is shown in Fig. 4. Four other less precise measurements were also made of the 4.4-minute activation curve. These auxiliary measurements

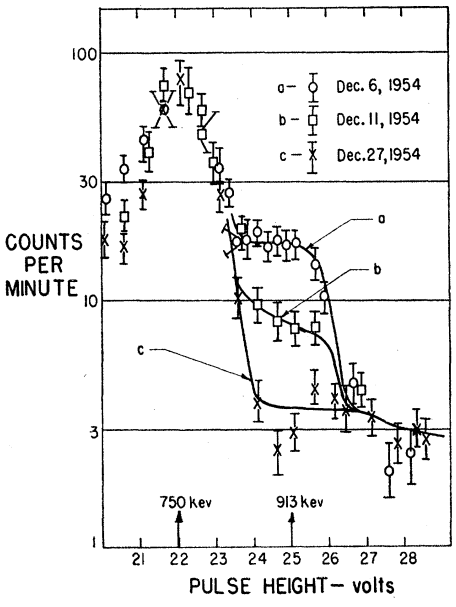

FIG. 3. Pulse-height spectrum of Zr^{89}.

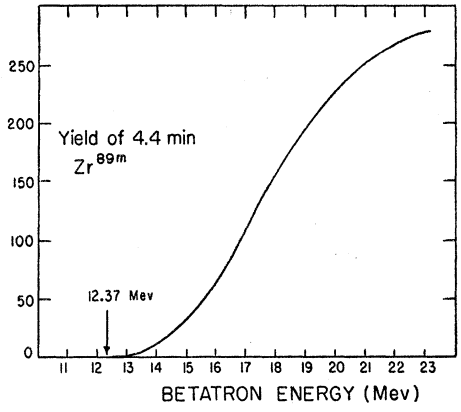

FIG. 4. 4.4-minute Zr^{89m} activation function.

were taken earlier and are mentioned because they gave consistent results even though they used rather different techniques. Two of these earlier measurements were taken with metal foils, a Victoreen monitor, and Geiger counter detection. The other two corroborative measurements were taken by using ZrO_2 disks, gamma-ray counting, and the ionization chamber monitor. When the ZrO_2 disks were used for 4.4-minute yield determination, the effect of the $O^{16}(\gamma,n)O^{15}$ reaction leading to the 2-minute positron emitter was subtracted.

The activation function for the 79-hour activity was determined precisely on two separate occasions by using ZrO_2 disks and counting the gamma rays in the 913-kev photopeak. One of these activation curves is shown in Fig. 5. Typical bombardment times, irradiation intensities, and counting rates are shown in Table I. Three other less precise sets of measurements were made using both ZrO_2 disks and Zr foils, both Victoreen and ionization chamber monitoring, and using both Geiger counter and scintillation detectors. As in the case of the 4.4-minute measurements, these auxiliary measurements showed that no gross errors were introduced by the more precise experimental procedure.

The 4.4-minute and the 79-hour activation curves and the cross sections extracted from them will be discussed further in Sec. VI.

IV. ABSORPTION MECHANISM AND COMPETITION BETWEEN NEUTRON AND GAMMA-RAY EMISSION

This section contains a quantitative comparison of the experimental yields with those predicted by theory. The theoretical yields are found by combining the calculated cross section with the calculated bremsstrahlung spectrum. Both the multipolarity of the absorbed gamma rays and the neutron gamma-ray competition contain adjustable parameters which make it possible to fit the experimental yield. Uncertainty is introduced into the calculated cross section by the model-dependent transmission coefficients which are used. In order to illustrate the type of information

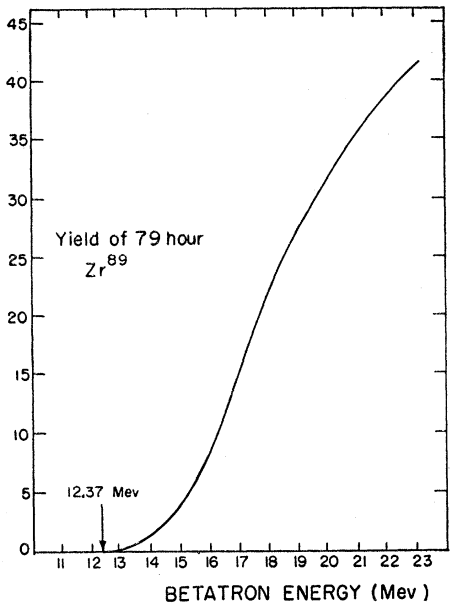

FIG. 5. 79-Hour Zr^{89} activation function.

which could be obtained if accurate transmission coefficients were available, the analysis is carried out as though the transmission coefficients were exact.

The analysis shows that the strong-interaction transmission coefficients have the correct energy dependences and the correct relative magnitudes to explain the experimental results. Even if these transmission coefficients are not exact, the experimental data require that the actual coefficients be similar to those used. On the other hand, if the strong-interaction transmissions are correct, the analysis indicates that electric dipole absorption is not dominant at energies of about 12.5 Mev. The maximum fraction of $E1$ absorption consistent with the experimental data varies from about 0.60 to 0.25 depending on the actual value of the competition between neutron and gamma-ray emission. Furthermore, if there is an appreciable amount of $M1$ absorption, the maximum admissable fraction of $E1$ absorption is even smaller.

(A) Description of Calculations

1. Compound Nucleus Hypothesis

The starting point of the theoretical analysis is the assumption that the (γ,n) process consists of gamma-ray absorption followed by the independent process of neutron emission, so that the cross section can be written as

$$\sigma_{\gamma,n}(E)=\sigma_a(E)P_n(E), \tag{1}$$

where σ_a is the absorption cross section for a gamma ray of energy E, and P_n is the probability of neutron emission.

The principal energy dependence of $\sigma_{\gamma,n}$ in the threshold region comes from $P_n(E)$, which actually depends on $\epsilon\equiv E-E_{\text{th}}$; in the energy region of interest ϵ varies from 0 to 1150 kev whereas E varies by only 10%. For Zr^{90}, the main competing mode of decay is gamma-ray emission, so that $P_n(\epsilon)$ can be written

$$P_n(\epsilon)=\frac{\Gamma_n(\epsilon)}{\Gamma_\gamma(E)+\Gamma_n(\epsilon)}, \tag{2}$$

where Γ_n is the partial width for neutron emission. Once again the main energy dependence comes from the factor depending on ϵ (i.e., Γ_n) rather than from the factor depending on E.

(The small proton emission despite the fact that the (γ,p) threshold is 3.29 Mev[18] below the (γ,n) threshold is due to the large Coulomb barrier. Calculations were made of the transmission of protons through the Coulomb barrier using standard penetrability tables,[21] and underestimating the Coulomb barrier by using a nuclear radius of $1.5\ A^{1/3}\times10^{-13}$ cm. The proton transmission 575 kev above the 79-hour threshold was only 0.0033 as opposed to a gamma-ray emission corresponding to at least 0.08. At 1150 kev above threshold, the proton transmission has increased to about 0.015 but the combined neutron and gamma-ray emission corresponds to at least 0.4 at this energy.

Experimental confirmation of the low proton emission rate was obtained by finding the yield of the $Zr^{90}(\gamma,p)Y^{89m}$ 13-second, 913-kev isomer at a betatron energy of 22 Mev. This (γ,p) yield was only 0.12% of the (γ,n) yield at the same energy.

A particularly careful search was made for the 13-second activity at betatron energies below 16 Mev but no 13-second 913-kev gamma rays could be observed. This intense search for the 13-second activity was made in an attempt to identify an observed gamma-ray activity of about 230-kev and 18-second

TABLE I. 79-Hour activation data.

Integrator setting	Energy (Mev)	Counting rate at end of bombardment in 913-kev photopeak (counts/min)	Bombarding time (min)	Ionization chamber current (μa)	Calculated dose in r at 50 cm	Count/min per 100 r
2200	13.30	172	144	285	7478	2.30
2300	13.92	635	260	441	11 572	5.49
2400	14.55	1875	235	707	18 552	10.10
2800	17.13	7552	156	515	13 514	55.89
3200	19.87	2905	15	101	2650	109.62
3600	22.82	21 408	100	591	15 508	138.04

[21] Feshbach, Shapiro, and Weisskopf, Atomic Energy Commission Report NYO 3077 (unpublished).

TABLE II. Typical transmission coefficients. (Reasonable assumptions about Γ_γ and Γ_n indicate that for nuclei of different A and E_{th}, $\Gamma_\gamma=\Gamma_n$ for T between about 0.004 and 1.0; for Zr^{90}, $\Gamma_\gamma=\Gamma_n$ for $T=0.8$.)

ϵ	1 ev	1 kev	10 kev	100 kev	300 kev	500 kev	1150 kev
$T_0(\epsilon)$	0.00078	0.027	0.085	0.245	0.362	0.434	0.58
$T_1(\epsilon)$		0.00004	0.0016	0.042	0.156	0.245	0.45
$T_2(\epsilon)$			0.0000032	0.00096	0.011	0.032	0.144
$T_3(\epsilon)$				0.000007	0.00024	0.0012	0.016

half-life. We are indebted to Dr. A. W. Sunyar of Brookhaven National Laboratory who aided us in identifying this activity by emphasizing to us the huge neutron capture cross section of Hf^{178}. The observed activity was subsequently shown to be identical with the well-known 19-second isomer in Hf^{179} by using small quantities of pure Hf for comparison. No other activities were observed from the Hf impurity in Zr, and the 19-second activity did not interfere with the Zr measurements.)

2. Introduction of Transmission Coefficients

In order to express $\Gamma_n(\epsilon)$ in terms of transmission coefficients,[1] it is convenient to write

$$\Gamma_n(\epsilon)=\frac{T_l(\epsilon)}{T_0(1)}\Gamma_n{}^0, \tag{3}$$

where $T_l(\epsilon)$ is the transmission of a neutron of energy ϵ, and angular momentum, $l\hbar$, through its centrifugal barrier; $T_0(1)$ is the transmission of an $l=0$ neutron of 1-ev energy, and $\Gamma_n{}^0$ is the (reduced) neutron width of a 1-ev neutron with $l=0$. The value of $\Gamma_n{}^0$ is expected to be proportional to the spacing, $D_{J\pm}$, of the levels of spin-parity, $J\pm$.[22] The symbol, $\Gamma_{J\pm}{}^0$, will be used to represent the reduced neutron width of the appropriate level.

The angular momentum, $l\hbar$, carried away by a neutron which leaves Zr^{89} in its 9/2+ ground state depends on the spin parity of the excited state in Zr^{90}. Electric dipole ($E1$) absorption leads to a 1− excited state and conservation of angular momentum and parity would require neutrons with $l=3$ (or $l=5$) to leave Zr^{89} in its ground state. Similarly, $E2$ absorption would require $l=2$ (or $l=4$ or $l=6$) neutrons and $M1$ absorption would require $l=4$ (or $l=6$) neutrons. For each mode of absorption, the higher l values given in the parenthesis are allowed by conservation rules but are unimportant because the transmissions, T_l, decrease rapidly with ϵ in the energy range of interest as shown in Table II. A rigorous treatment would specify the neutron transmission from the 2+ level to the 9/2+ level as

$$T_{2+\to 9/2+}(\epsilon)=g_2T_2(\epsilon)+g_4T_4(\epsilon)+g_6T_6(\epsilon), \tag{4}$$

where g is a statistical weight factor which takes into account the vectorial addition of 2+ and l, to form the neutron spin and the 9/2+ Zr^{89} spin. It is a satisfactory approximation to neglect higher l values and use only the leading term. Setting the g-factor equal to 1, Eq. (4), which applies to $E2$ absorption, becomes

$$T_{2+\to 9/2+}(\epsilon)=T_2(\epsilon). \tag{5}$$

The theoretical (γ,n) cross section depends on the fraction of gamma rays, f, absorbed in an $E2$ process and the fraction f' absorbed in an $M1$ process. If higher multipoles are unimportant, the fraction absorbed in an $E1$ process is $(1-f-f')$.

Using these definitions and assumptions, Eqs. (1)–(5) can be combined to give the (γ,n) cross section for $\epsilon\lesssim 575$ kev:

$$\sigma_{\gamma,n}(E)=\sigma_a(E)\left\{\frac{fT_2(\epsilon)}{r+T_2(\epsilon)}+\frac{(1-f-f')T_3(\epsilon)\gamma_{1-,2+}}{r+T_3(\epsilon)\gamma_{1-,2+}}+\frac{f'T_4(\epsilon)\gamma_{1+,2+}}{r+T_4(\epsilon)\gamma_{1+,2+}}\right\}, \tag{6}$$

where

$$r\equiv\Gamma_\gamma T_0(1)/\Gamma_{2+}{}^0,\quad \gamma_{1-,2+}\equiv\Gamma_{1-}{}^0/\Gamma_{2+}{}^0,$$

and

$$\gamma_{1+,2+}\equiv\Gamma_{1+}{}^0/\Gamma_{2+}{}^0.$$

It is fortunate that the final results are not particularly sensitive to $\gamma_{1-,2+}$ and $\gamma_{1+,2+}$ since these ratios are not known, even though they are expected to be near 1. One would expect[22] $\gamma_{1-,2+}\cong D_{1-}/D_{2+}$ and $\gamma_{1+,2+}\cong D_{1+}/D_{2+}$ but the relative level spacings of different spin states is unknown. Hughes, Garth, and Levin[23] were unable to detect any variation of D_J with J, thus implying that $\gamma_{J,J'}=1$. On the other hand, some theoretical papers[24–26] use the estimate that $D_J\propto(2J+1)^{-1}$ which would make $\gamma_{1+,2+}=5/3$. For simplicity, we use $\gamma_{1,2}=1$.

3. Estimates of Competition

The value of r in Eq. (6) is also uncertain but it is kept as a parameter and we express f as a function of r. Values of r have been measured by Harvey, Hughes, Carter, and Pilcher[27] who examined neutron resonances throughout the periodic table. In the neighboring element, Mo, they found $\Gamma_\gamma=(260\pm80)\times10^{-3}$ ev, and for the even-even compound nuclei of Mo^{96} and Mo^{98} the average of $\Gamma_n{}^0$ was about 13×10^{-3} ev. Since $T_0(1)$

[22] See for example, Blatt and Weisskopf, reference 1, p. 389 ff and p. 420 ff.

[23] Hughes, Garth, and Levin, Phys. Rev. **91**, 1423 (1953).
[24] L. Wolfenstein, Phys. Rev. **82**, 690 (1951).
[25] W. Hauser and H. Feshbach, Phys. Rev. **87**, 366 (1952).
[26] J. M. B. Lang and K. J. Le Couteur, Proc. Phys. Soc. (London) **A67**, 586 (1954).
[27] Harvey, Hughes, Carter, and Pilcher, Phys. Rev. **99**, 10 (1955).

$=8\times10^{-4}$, these values correspond to an r of about 0.016. However, since r is a function of energy, additional data are necessary in order to estimate r for Zr^{90} since it has a much higher (γ,n) threshold than either 9.15 or 8.3 Mev which are the values of Mo^{96} and Mo^{98}, respectively. An indication of the variation of r with energy can be obtained by using the theoretical prediction that Γ_n^0/D is a constant or at least relatively independent of energy.[22] Since Γ_γ varies comparatively slowly with energy, the energy dependence of r would be governed by D which decreases exponentially with energy. Harvey, Hughes, Carter, and Pilcher[27] have experimentally verified the relative constancy of Γ_n^0/D and find that this ratio is 4×10^{-5} for Mo. The variation of D with energy was taken from the formula of Lang and Le Couteur[26] which agrees fairly well with the measurements of Harvey *et al.*[27] The calculated value of r corresponding to the (γ,n) threshold value of Zr^{90} is $r=0.8$ when one uses measured Γ_n^0/D, the measured Γ_γ, and the empirically fitted formula for D. This value of r is based on a predicted D of 6.5 ev and therefore a Γ_n^0 of 0.26×10^{-3} ev. This estimate of r is only a coarse approximation and does not take into account either the possible increase in Γ_γ that might be expected with higher E or the larger value of D that might be expected because Zr^{90} has a closed shell of 50 neutrons. It should be mentioned that for most other nuclei r would be considerably smaller than 0.8. A hypothetical isotope of Zr with a threshold of 6 Mev would have an r of about 0.004. For typical heavier nuclei, r would be about 0.03.

4. *Effect of Excited States*

The (γ,n) cross section of Zr^{90} is given by Eq. (6) and is correct only for excitation energies insufficient to leave Zr^{89} in its first excited state, presumably Zr^{89m}. Above the threshold for the production of 4.4-minute Zr^{89m}, neutrons with an energy $\epsilon'=E-E_{\text{th}}-0.59$ Mev can leave Zr^{89} in this excited state and thereby change the yield of the 79-hour activity in two ways. These competing neutrons would reduce the direct production of the 79-hour activity below what would be expected from Eq. (6). On the other hand, the 79-hour activity would grow indirectly through the 4.4-minute isomeric state.

In order for Zr^{89} to be left in its $\frac{1}{2}-$ first excited state $l=0$ neutrons would have to be emitted in the case of $E1$ absorption and $l=1$ neutrons would have to be emitted for either $E2$ or $M1$ absorption. In these three cases, neutrons with higher l values (i.e., $l=2$, $l=3$, and $l=3$, respectively) are once again neglected. The reduction in the direct production of the 9/2+ state due to competition appears theoretically in the form of an addition term in the denomintor of each term in Eq. (6). As an example, the term corresponding to $E1$ absorption becomes

$$\sigma_{\gamma,n}(E)_{\text{79-hour, direct, }E1}=\sigma_a(E)\frac{(1-f-f')T_3(\epsilon)\gamma_{1-,2+}}{r+[T_3(\epsilon)+T_0(\epsilon')]\gamma_{1-,2+}}. \quad (7)$$

The denominators of the terms corresponding to $E2$ and $M1$ absorption in Eq. (6) would both add $T_1(\epsilon')$ as a term in a similar manner [and become equal to the denominators in Eq. (9)].

The indirect production of the 79-hour activity through the 4.4-minute state occurs in 93% of the 4.4-minute decays[5]:

$$\sigma_{\gamma,n}(E)_{\text{79-hour, indirect}}=0.93\sigma_{(\gamma,n)}(E)_{\text{4.4-minute}}. \quad (8)$$

The 4.4-minute cross section can be written as

$$\sigma_{\gamma,n}(E)_{\text{4.4-min}}=\sigma_a(E)\left\{\frac{fT_1(\epsilon')}{r+T_2(\epsilon)+T_1(\epsilon')}+\frac{(1-f-f')T_0(\epsilon')\gamma_{1-,2+}}{r+[T_3(\epsilon)+T_0(\epsilon')]\gamma_{1-,2+}}+\frac{f'T_1(\epsilon')\gamma_{1+,2+}}{r+[T_1(\epsilon')+T_4(\epsilon)]\gamma_{1+,2+}}\right\}. \quad (9)$$

5. *Accuracy of Transmission Coefficients*

The transmission coefficients for Eqs. (6)–(9) were calculated from the standard formulas[1] which use an ordinary (real) potential well whose depth is such that a zero-energy neutron which penetrates into the nucleus has a wave number, $K_0=10^{-13}$ cm.[28] These transmission coefficients are possibly incorrect since they are based on the "strong-interaction" model. Feshbach, Porter, and Weisskopf have more recently proposed a "cloudy crystal ball" or "optical" model which uses a potential containing a small imaginary term.[29] According to some sample calculations done by Dr. Sophie Oleksa at Brookhaven National Laboratory, the optical model in its present form has a radical effect on transmission coefficients at some energies and in some parts of the periodic table.[30,31] (The optical model transmission coefficients necessary to calculate the (γ,n) cross section for Zr^{90} were not yet available when this paper was written.) Although the strong-interaction transmission coefficients are probably somewhat incorrect, they may be closer to the actual coefficients than those given by the present form of the optical model.[30] The older transmission factors have been used (even after the

[28] J. Blatt and V. F. Weisskopf, reference 1, p. 355. This presentation follows the treatment given by H. Feshbach and V. F. Weisskopf, Phys. Rev. **76**, 1550 (1949).

[29] Feshbach, Porter, and Weisskopf, Phys. Rev. **96**, 448 (1954). For earlier suggestions of the optical model see H. A. Bethe, Phys. Rev. **57**, 1125 (1940), and Fernbach, Serber, and Taylor, Phys. Rev. **75**, 1352 (1949).

[30] Reported by H. Feshbach, Brookhaven National Laboratory Report BNL 331(C21), p. 61 ff (unpublished).

[31] C. E. Porter, (private communication).

TABLE III. Some typical values of σ_l/σ_a as calculated from Eqs. (6), (7), and (9) with $f'=0$.

		ϵ for $l=2$ and 3; ϵ' for $l=0$ and 1.				
		50 kev	100 kev	300 kev	500 kev	1150 kev
$f=0$	$l=0$ $r=0.08$	6.8×10^{-1}	7.5×10^{-1}	8.0×10^{-1}	8.25×10^{-1}	
	$l=0$ $r=0.8$	1.44×10^{-1}	2.15×10^{-1}	3.0×10^{-1}	3.4×10^{-1}	
$f=1$	$l=1$ $r=0.08$	1.0×10^{-1}	2.4×10^{-1}	4.8×10^{-1}	5.37×10^{-1}	
	$l=1$ $r=0.8$	1.75×10^{-2}	4.6×10^{-2}	1.49×10^{-1}	2.08×10^{-1}	
$f=1$	$l=2$ $r=0.08$	2.1×10^{-3}	1.19×10^{-2}	1.21×10^{-1}	2.8×10^{-1}	2.91×10^{-1}
	$l=2$ $r=0.8$	2.1×10^{-4}	1.2×10^{-3}	1.4×10^{-2}	3.8×10^{-2}	1.18×10^{-1}
$f=0$	$l=3$ $r=0.08$	1.0×10^{-5}	8.8×10^{-5}	3.0×10^{-3}	1.47×10^{-2}	2.98×10^{-2}
	$l=3$ $r=0.8$	1.0×10^{-6}	8.8×10^{-6}	3.0×10^{-4}	1.47×10^{-3}	1.28×10^{-2}

advent of the new model) with considerable success in interpreting inelastic neutron scattering experiments[30,32] according to the theory of Hauser and Feshbach.[25]

6. Calculation of Cross Sections

The cross sections for the formation of the 79-hour activity and the 4.4-minute activity were calculated for various values of r, for $\sigma_a(E)$ constant, for $f'=0$ and for $f=1$ or $f=0$. Both the direct and indirect formation of the 79-hour activity were included. The effects of $\sigma_a(E)$ varying with E, of $f'\neq0$, and of $0<f<1$ were easily investigated after the yields of the activities had been calculated. Typical values of σ_l/σ_a are shown in Table III.

7. Calculation of Yields

The yields of the activities at different betatron energies, E_β, were computed from these cross sections by numerical integration of Eq. (10):

$$Y_j(E_\beta)=\int_{E_{\rm th}}^{E_\beta} N(E,E_\beta)\sigma_j(E)dE, \qquad (10)$$

where $N(E,E_\beta)dE$ is the calculated number of gamma rays in the energy range between E and $E+dE$ formed by electrons of energy, E_β [i.e., $N(E,E_\beta)$ is the bremsstrahlung photon spectrum]. The spectrum had been calculated by Leiss and Penfold[33] using the formula derived by Schiff[34] for bremsstrahlung x-rays emitted in the forward direction and for a thin target. C was set equal to 191 in Schiff's formula. The spectrum, which was available at 5-kev intervals, was converted into the number of photons in 25-kev bins for the numerical integration. Since the calculated spectra were normalized per incident electron on the betatron target, the calculated yield, $Y(E_\beta)$, was divided by E_β to compensate for the monitor response. This first order correction for the monitor response never exceeded 5 percent and a more refined correction was therefore unwarranted. An exaggerated thick-target spectrum was used for some calculations and it produced relatively unimportant changes in the quantitative results and did not affect the general conclusions. [The thick-target spectrum was generated by assuming that at a betatron setting of E_β there were actually 20 equally probable energies of the bremsstrahlung producing electrons and that these 20 energies varied in 5-kev steps from $E_\beta-95$ kev to E_β. The actual effect of the target thickness on the spectrum produced by the Illinois betatron has been shown to be smaller than this by Penfold and Spicer in their careful work on the fine structure in the $O^{16}(\gamma,n)O^{15}$ reaction.[35]] Typical values of the yields calculated from Eq. (10) by using the thin target spectrum are given in Table IV for $E_\beta-E_{\rm th}=575$ kev and 1150 kev.

(B) Results—Comparison between Calculated and Experimental Values

The three experimental results which can be analyzed to give information about the absorption mechanism include: (1) the energy dependence of the 79-hour activity between $E_{\rm th}+375$ kev and $E_{\rm th}+575$ kev, (2) the energy dependence of the 4.4-minute activity up to 600 kev above its threshold, and (3) the ratio of the yield of the 79-hour activity at 575 kev to that at 1150 kev above threshold.

(1) The yield of the 79-hour activity for the three values of betatron energy E_β, 375 kev, 475 kev, and 575 kev above the 79-hour threshold energy $E_{\rm th}$, can be fitted empirically by

$$Y(E_\beta)\propto(E_\beta-E_{\rm th})^m, \quad m=3.4\pm0.8. \qquad (11)$$

The corresponding variation of cross section with energy is

$$\sigma(E)\propto(E_\beta-E_{\rm th})^p, \quad p=2.1\pm0.7. \qquad (12)$$

This energy dependence corresponds to that expected for $l=2$ neutrons if the strong-interaction transmission coefficients are correct. Since $Y_3(575)\ll Y_2(575)$, the yield at energies up to 575 kev above threshold is determined almost completely by $Y_2(\epsilon)$ provided only that $f\geq0.1$ (see Table IV). $Y_4(\epsilon)$, which would arise from $M1$ absorption, is negligibly small. $T_2(\epsilon)\propto\epsilon^{2.1}$ for 100 kev$<\epsilon<$600 kev and thus $\sigma_{\gamma,n}\propto\epsilon^{2.1}$ if r is much larger than $T_2(\epsilon)$. If r is relatively small (but larger than the minimum of 0.08 established below), the

[32] Bernard Margolis, Columbia University, verbal report, Medium Energy Neutron Conference at Argonne National Laboratory, June 3, 1955 (unpublished).
[33] J. E. Leiss and A. S. Penfold (unpublished).
[34] L. I. Schiff, Phys. Rev. 83, 252 (1951).
[35] A. S. Penfold and B. M. Spicer (to be published and private communication).

TABLE IV. Typical values of the calculated yield of 79-hour activity. (Note: If $\sigma_{\gamma,n}/\sigma_{abs}=1$, the yield would be 7060 for ϵ or $\epsilon'=575$ Kev, 20 400 for $\epsilon=1150$ kev.)

$r=\Gamma_\gamma T_0(1)/\Gamma_n^0$	Direct $f=1$ $Y_2(575)$	Direct $f+f'=0$ $Y_3(575)$	Direct $f=1$ $Y_2(1150)$	Direct $f+f'=0$ $Y_3(1150)$	Indirect $f=1$ $0.93Y_1(575')$	Indirect $f+f'=0$ $0.93Y_0(575')$
$r=0.08$	705	26.1	3620	164	2230	4500
$r=0.8$	87	3.19	733	22	679	1700

calculated $\sigma_{\gamma,n}(\epsilon)\propto\epsilon^{1.7}$. Thus, for the entire acceptable range of r, the governing $l=2$ term has the correct energy dependence. In contrast, $l=3$ neutrons would have too rapid an energy dependence (even for the smallest admissible r value) and $l=1$ neutrons have too slow an energy dependence (even for very large r values). The experimental data are not sufficiently precise to eliminate the possibility of admixtures of $l=1$ or $l=3$ neutrons; however, the dominant group of emitted neutrons has $l=2$. On the other hand, since $Y_1\gg Y_2$, a small amount of $E3$ absorption would tend to make $l=1$ neutrons dominant in contradiction with the experimental results.

If the strong-interaction transmission factors are not correct, the correct transmission factor for the dominant neutron group must have the same energy dependence as does the $l=2$, strong-coupling transmission factor. It is entirely possible that the actual transmission factors are radically different from the strong-coupling factors but give the misleadingly correct energy dependence. Although this is unlikely, the quantitative results derived using theoretical transmission factors to interpret the experiment must be considered tentative until the correct transmission factors are known.

(2) The yield of the isomeric level, $Y(E_\beta)_{4.4\text{-min}}$ for 600 kev above its threshold, E_{th}', can also be fitted by a power law:

$$Y(E_\beta)_{4.4\text{-min}}\propto(E_\beta-E_{th}')^n,\quad n=1.95\pm0.2. \qquad (13)$$

This rapid variation of yield with energy requires explanation since if neutron emission predominated over gamma-ray emission for $\epsilon>10$ kev (as might be expected), the (γ,n) cross section would be essentially

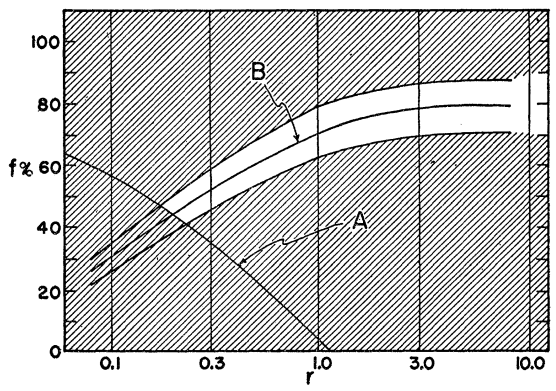

FIG. 6. Allowable values of f and r. f is the fraction of $E2$ absorption if there is no $M1$ absorption. $r=8\times10^{-4}\Gamma_\gamma/\Gamma_n^0$.

independent of energy [Eqs. (1) and (2)], and n would be equal to only about 1.5. The implied relative improbability of neutron emission indicates either that r is large (in which case the observed energy dependence could be explained even for $E1$ absorption and $l=0$ neutrons) or that $E2$ and $M1$ absorption are important (in which case the rapid energy dependence could be explained by the transmission $T_1(\epsilon')$ of the $l=1$ neutrons). Thus, if r is small, $f+f'$ must be large whereas if r is large, $f+f'$ is not restricted and could be quite small. The actual restriction placed on f and r is shown by curve A in Fig. 6 and by the excluded (shaded) area to the left of curve A. Curve A was calculated by matching Eqs. (9) and (10) with $n=1.75$, the smallest n value consistent with the measurements. The ordinate, f, on Fig. 6 is appropriate for curve A only if $\gamma_{1-,2+}=\gamma_{1+,2+}=1$ and $f'=0$; the proper ordinate for curve A is $(f+f'\gamma_{1+,2+})\gamma_{1-,2+}$.

Curve A does not appear to be particularly restrictive for Zr^{90} because r is expected to be relatively large. However, there are many nuclei for which n is close to 2.0[13,16] and for which r is expected to be about 0.03, (according to Sec. IV.A.3). For these nuclei, curves like curve A are quite informative. It can be interpreted to indicate either relatively large amounts of each type ($E2$ and $M1$) absorption so that there would be an appreciable fraction of $l=1$ neutrons or anomalously large r values as might be expected if the states reached by gamma-ray absorption emitted gamma rays preferentially.

(3) The most restrictive data on f obtained with Zr^{90} came from the ratio of the 79-hour yield at 575 kev to that 1150 kev above the 79-hour threshold. The experimental value of this ratio, $Y(575)/Y(1150)$ was found to be 0.04 ± 0.005. Theoretical values were calculated for values of r between 0.025 and 12 and for all values of f and f'.

If the 4.4-minute isomeric level were unknown, the measurements at these two energies might be thought to imply a rapid energy dependence of the yield, i.e., $Y(E_\beta)\propto(E_\beta-E_{th})^{4.6}$. Actually, as is shown in Table IV, the 79-hour yield above the 4.4-minute threshold energy is largely the result of indirect production which is proportional to $(E_\beta-E_{th}-0.59\text{ Mev})^2$ rather than to $(E_\beta-E_{th})^n$. Thus, whereas the ratio of 0.04 might appear anomalously low if there were no indirect production, the importance of the indirect production makes this ratio rather large.

The direct production observed at 575 kev is too

large compared with the total production at 1150 kev to be explained by the emission $l=3$ neutrons. Since there is no value of r for which $Y_3(575)$ is big enough to explain the experimental ratio, most of the observed yield at 575 kev must be due to $Y_2(575)$ (i.e., $l=2$ neutrons) arising from $E2$ absorption. On the other hand, pure $E2$ absorption (i.e., $f=1$) predicts too large a ratio; $Y_2(575)/[Y_2(1150)+0.93Y_1(575')]$ is greater than 0.04 for all values of r. However, this theoretical ratio can be reduced to 0.04 by reducing f below 1, which decreases the theoretically predicted numerator and increases the denominator. The numerator would be reduced essentially by a factor of f since both $f'Y_4$ and $(1-f-f')Y_3$ are negligible. Reducing f below 1.0 adds a term to the denominator which is

$$(1-f)[0.93Y_0(575')-0.93Y_1(575') +Y_3(1150)-Y_2(1150)].$$

The actual value of f necessary to fit the experimental ratio depends on r—for small values of r, f must be reduced more than would be necessary for large r values in order to match experiment. (If r were very large, $Y_l(E_\beta)$ would be proportional to $1/r$; for nominal values as r decreases $Y_l(E_\beta)$ does not increase as much for small values of l as it does for large values of l.)

The values of f for different r values (and for $f'=0$) implied by $Y(575)/Y(1150)=0.04$ are shown by curve B in Fig. 6. The extreme values of f consistent with the limits of the experimental errors are shown by the curves on each side of B. The excluded region of the f–r plane is shaded. The particularly interesting feature of Fig. 6 is the large value of f which will be discussed further after the effect of $M1$ absorption is given below.

$M1$ absorption decreases both the $E2$ and the $E1$ absorption below the values appropriate for no $M1$ absorption; i.e., if $f'\neq 0$, both f and $(1-f-f')$ decrease. This effect can be understood qualitatively inasmuch as a finite f' would not affect the yield at 575 kev (provided r and f remained the same) but it would decrease the indirect yield at 1150 kev by subtracting the term, $0.93f'[Y_0(575)-Y_1(575)]$. Thus $f'\neq 0$ would tend to make the calculated yield ratio too large and f would have to be reduced in order to fit the experimental value. For small r values the reductions in f (due to $f'\neq 0$) are very small because $Y_0(575')$ is only slightly larger than $Y_1(575')$. For larger r values, $\Delta f=-1/2\Delta f'$. Table V shows the fractional values of $E2$, $M1$, and $E1$ absorption, i.e., f, f', and $(1-f-f')$ for four values of r.

It would be possible to construct a set of curves corresponding to Fig. 6 (which applies to $f'=0$) for each value of f'. Curve A would shift downwards on these new graphs since $f+f'$ should be the ordinate for curve A. Just as $r=0.17$ is the minimum admissible r value defined by curves A and B for $f'=0$ in Fig. 6, the minimum r values for $f'=0.2$ and 0.4 would be 0.11 and 0.065, respectively.

Since f would be increased by any change which decreases the theoretical ratio of $Y(575)/Y(1150)$, the effect of several of the approximations can be estimated. If the effective target thickness were appreciable, the yield at 575 kev would be decreased proportionately more than the yield at 1150 kev and the necessary value of f would be increased. The exaggerated thick target spectrum increases f by about 15%; therefore the actual effect of target thickness probably increases f by only a few percent. Another possible change which might increase f would be an increase in the gamma-ray absorption cross section, σ_a, with energy. It is quite likely that σ_a increases with E but the exact energy dependence is difficult to estimate. A probable extreme of the energy dependence would be $\sigma_a \propto E^3$; and even this rapid energy dependence would increase f by only 15%. A refinement of the assumption that r is a constant would tend to decrease f somewhat. As the energy increases, r would increase because D and therefore Γ_n^0 would decrease. In going from an excitation of 12 Mev to 12.5 Mev in Zr^{90}, D would be expected[26] to decrease by a factor 0.68; if Γ_γ does not increase appreciably, r would therefore increast by a factor of 1.5 for the higher energy. The exace effect on f of an increase of r with energy would depend on the specific values of r, f, and f'. For small values of r, the yield at 1150 kev is quite insensitive to r and therefore f is not affected much. For larger values of r, f would tend to decrease by as much as 15%.

(C) Conclusions

Even though this experiment does not give explicit values for r, f, and f', it is possible to reach some interesting conclusions based on the interpretation of the results.

(1) This experiment shows directly that $E1$ absorption is somewhat less important at low energy that it is at higher energies. Electrodisintegration experiments

TABLE V. Fractional amounts of $E2$, $M1$, and $E1$ absorption.

Fraction of $M1=f'$	$r=0.08$		$r=0.24$		$r=0.8$		$r=2.4$	
	f	$1-f-f'$	f	$1-f-f'$	f	$1-f-f'$	f	$1-f-f'$
0	0.25	0.75	0.48	0.52	0.68	0.32	0.78	0.22
0.2	0.237	0.56	0.43	0.37	0.60	0.20	0.68	0.12
0.4	0.224	0.38	0.39	0.21	0.52	0.08	0.58	0.02
0.6	0.210	0.19	0.34	0.06				
0.8	0.197	0.0						

at higher energies were consistent[36] with a fractional $E1$ absorption (i.e., $1-f-f'$) of 0.88.

This experiment sets an upper limit of about 0.60 on $1-f-f'$. The reduced importance of $E1$ absorption at low energy is consistent with expectations of theory which predicts that below the "giant resonance" $M1$ absorption would be somewhat larger than $E2$ absorption and that both would be larger than $E1$ absorption.[37]

(2) If the approximate value of $r=0.8$ is accepted (from quite independent nuclear data as shown in Sec. IV.A.3), then $1-f-f'$ has a maximum value of 0.39 for $f'=0$. The maximum value that f' could have for this r value would be 0.6 in which case $f=0.4$ and $E1$ absorption would be 0. This set of values ($f'=0.6$, $f=0.4$, $r=0.8$) agrees both with expected r values and the expected dominance of $M1$ absorption.

(3) If $M1$ absorption were required to be appreciably larger than $E2$ absorption,[37] only smaller r values would be acceptable as shown in Table V. If $f'\geq 2f, r\leq 0.24$; for r as small as 0.08, f' could be about five times f.

(4) Although no firm upper limit can be placed on r, there are two evidences that r is not much larger than the predicted value of 0.8. First, a much larger r value would imply that the observed (γ,n) cross section would only be a small fraction of the absorption cross section until the energy is high enough to reach many levels in Zr^{89}. Since the observed (γ,n) cross section has a reasonable shape, it seems unlikely that σ_a is radically different at low energies. The second evidence for r not being much larger than 0.8 comes from the relative values of f' and f. If one kept the restriction that $f'>f$, r would be confined to low values. This restriction may not be valid if there are particularly few 1+ levels in Zr^{90} compared with 2+ levels. However, if the level density does not differ by more than a factor of 2 or 3, f' would not be smaller than f and r would be restricted to less than 1.5.

A restriction on the maximum value of r would be quite significant since it would tend to confirm the validity of the compound nucleus in (γ,n) reactions. If the absorbed gamma ray did not form a compound nuclear state, the excited state that was formed would be expected to be particularly likely to re-emit a gamma ray. Similarly one might expect that the excited states formed in neutron scattering experiments have a special tendency to re-emit neutrons. Thus, if the compound nucleus were not a valid model, the r values appropriate to (γ,n) reactions would be expected to be considerably larger than those derived from neutron scattering experiments.

The results obtained in this experiment are consistent with the compound nucleus theory and other assumptions made in Sec. IV.A. However, with our present knowledge, this experiment neither proves the assumptions nor fixes the values of the parameters. With additional similar experiments (e.g., some that were chosen to be particularly sensitive to $M1$ or $E1$ absorption), it should be possible to determine the absorption mechanism. Once this has been determined, photonuclear experiments of this type should give both values for r and the relative energy level densities for states of different spin parity.

V. THRESHOLD DETERMINATIONS

The determination of (γ,n) thresholds involves an extrapolation of the experimental data to the threshold energy because the yields at energies just above the threshold are in doubt due to both the small values of the yields and the non-negligible background effect. One extrapolation procedure which has been used with apparent success involves choosing values of both the threshold energy, E_{th}, and the energy dependence, m, so that the yield at a betatron energy of E_β, $Y(E_\beta)$, is given by[13,38,39]

$$Y(E_\beta)\propto(E_\beta-E_{\text{th}})^m. \tag{14}$$

It will be shown in this section that the apparent precision with which E_{th} can be determined from Eq. (14) is misleading and that there is little theoretical justification of this equation in most (γ,n) processes.

The energy dependence of $Y(E_\beta)$ is governed by the energy dependence of the (γ,n) cross section, $\sigma_{\gamma,n}(E)$, and on the bremsstrahlung spectrum $N(E,E_\beta)$ as indicated by Eq. (10). If both $\sigma_{\gamma,n}(E)$ and $N(E,E_\beta)$ had simple energy dependences, $Y(E_\beta)$ might be expected to satisfy Eq. (14). For example, if $\sigma\propto(E-E_{\text{th}})^p$ and $N(E,E_\beta)\propto(E_\beta-E)^q$, then it can be shown easily that $Y(E_\beta)$ satisfies Eq. (14) for the following simple cases:

(1) For $p=0$ or $p=1$, $m=p+q+1$ for all q,

(2) For $p=1/2$ and q an integer, $m=p+q+1$.

However, because neither p nor q is a constant, Eq. (14) does not hold. Rather than being represented by $q=$constant, the actual $N(E,E_\beta)$ corresponds to a value of q which is about 0.33 within 100 kev of the tip but which changes to about 0.56 for energies more than 200 kev from E_β. If the actual bremsstrahlung spectrum were used with a cross section that was a pure power law (i.e., $p=0$ or $p=1$), $Y(E_\beta)$ would be representable by Eq. (14) for energies $E\geq E_{\text{th}}+1$ Mev. At these higher energies, $m=p+1.6$. If one were to assume that Eq. (14) did hold for this case, one would choose a value of E_{th} which was about 80 kev above the actual threshold energy, even if precise experimental points were available within 200 kev of the actual threshold.

A further complication is introduced in an actual situation because the cross section is not a pure power law. Even if only one level in the residual nucleus were involved, the existence of $E2$, $M1$, and $E1$ absorption and the variation of the relevant transmissions would

[36] K. L. Brown and R. Wilson, Phys. Rev. **93**, 443 (1954).
[37] J. M. Blatt and V. F. Weisskopf, reference 1, p. 651 ff.
[38] Sher, Halpern, and Mann, Phys. Rev. **84**, 387 (1951).
[39] Halpern, Nathans, and Yergin, Phys. Rev. **95**, 1529 (1954).

cause a variation in the power law. Furthermore, when additional levels occur in the residual nucleus, the cross section changes. There are extreme cases in which the observed cross section might be due entirely to an excited state rather than the ground state,[40] thereby leading to large errors in neutron binding energies inferred from (γ,n) threshold measurements. Another reason for the (γ,n) cross section deviating from a simple power law is the variation in the photon absorption cross section.

Thus, although in some instances the combined effect of the energy variations of bremsstrahlung and cross section might compensate, the energy dependence of the yield cannot be assumed to be a simple power law as shown in Eq. (14). On the other hand, some extrapolation procedure is necessary in order to determine the threshold. The most satisfactory procedure would be to obtain the best points possible near threshold and then to plot $Y(E_\beta)^{1/m}$ as a function of E for several values of m. For example, when this procedure was used on the 4.4-minute threshold data, shown in Fig. 1, a value of $m=2$ seemed to fit best but $m=1.5$ and 2.5 were also tried. These different m values gave threshold energies 20 kev above and 20 kev below that given by the $m=2$ value. In contrast with this relatively small threshold shift, assuming Eq. (14) valid for more than 1 Mev would lead to thresholds 100 kev too high for $m=1.5$ and 80 kev too low for $m=2.5$.

There are two reasons which explain why the assumption of Eq. (14) and the adjustment of m and E_{th} from a log-log plot introduce more error than is introduced by plotting the mth root. First, the power, m, is determined essentially by experimental data in which $E_\beta > E_{\text{th}}+1$ Mev since the apparent value of m below 1 Mev is quite sensitive to the value of E_{th}. Thus, the log-log plot procedure assumes the constancy of m which is quite doubtful and obtains a misleadingly precise value of an apparent threshold. The second reason for the error introduced by this procedure is that there is a tendency to give very little weight to the most significant, low-energy points merely because they are least precise.[38] In contrast, using the plot of the mth root of $Y(E_\beta)$ *vs* E_β makes it easier to give these low-energy points the weight they deserve.

Sher, Halpern, and Mann[38] have shown that in some cases, assumption of a power law over a range of several Mev made it possible to obtain the correct thresholds for different isotopes of the same element. In view of the factors discussed above which would tend to change the power law, this observed agreement should probably be considered fortuitous and the procedure should not be assumed to have unwarranted reliability because of this agreement. Another apparent indication of the validity of the log-log plot was given by Birnbaum.[13] However, Birnbaum's data showed that Eq. (14) permitted a precise determination of an apparent threshold

[40] Axel, Fox, and Parker, Phys. Rev. **97**, 975 (1955).

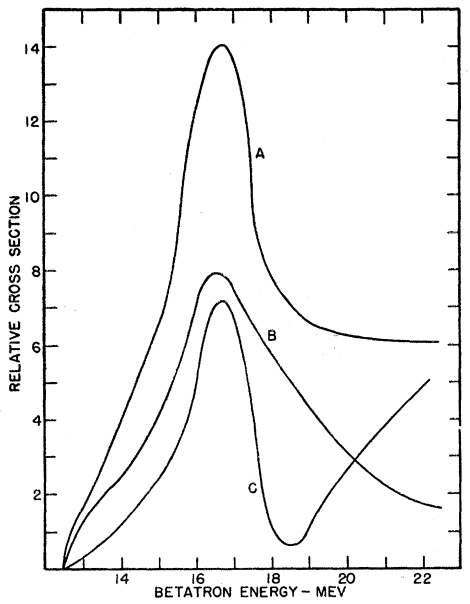

FIG. 7. Relative cross sections. Curve A is the cross section for the formation of the 79-hour activity, Curve B the formation cross section of the 4.4-minute activity, and Curve C the direct production cross section of the 79-hour activity.

E_{th}', but did not show that E_{th}' was an accurate threshold value. The constancy of m for a single isotope is difficult to reconcile with the theoretical considerations mentioned above, particularly when m has been found[38] to vary from element to element. For these reasons, and because the possible error introduced by using the wrong m is easier to ascertain, it is probably better to plot the mth root of $Y(E_\beta)$ *vs* E_β for determining thresholds. A superior extrapolation procedure can be expected only when the theory has been developed to the point where it can correctly predict the exact value of m or the variation of m with energy.

VI. INTERPRETATION OF THE YIELDS AT HIGHER ENERGIES

A. Calculation of the Cross Sections

The activation functions shown in Figs. 4 and 5 were converted into cross sections by using the inverted bremsstrahlung tables prepared by Penfold and Leiss[41] to perform a "photon-difference" analysis.[42] The relative cross section for the formation (direct and indirect) of the 79-hour Zr^{89} and the 4.4-minute Zr^{89m} are shown as curves A and B, respectively in Fig. 7. Since the 79-hour activity and the 4.4-minute activity were detected by counting gamma rays of different energy, the relative yield of the two activities at any betatron energy is in doubt. In order to provide maximum detection sensitivity, thick samples and "poor" counting geometry were used, thus making it difficult to calculate relative

[41] A. S. Penfold and J. E. Leiss (private communication).
[42] L. Katz and A. G. W. Cameron, Can. J. Phys. **29**, 51 (1951).

detection efficiencies. An independent measurement was therefore made in order to obtain the relative scales of curves A and B in Fig. 7. The normalization of the two cross sections was carried out through the normalization of the activation functions by two independent methods which give essentially the same result.

(1) Once the shapes of the activation functions had been determined, the relative yields of the Zr^{89} ground-state 913-kev and 588-kev isomeric transition gamma rays were compared using the 1 in.×1 in.×2 in. NaI(Tl) crystal and small sources suspended 2 in. from the 1 in.×1 in. face. With this "good" geometry, it was possible to calculate the relative efficiencies of the scintillation crystal for the 588-kev and 913-kev radiations. Samples of Zr metal weighing 200 mg were irradiated at 22.8 Mev and the activities were determined by using the apparatus described above. The ratio of the 79-hour 913-kev yield to the yield of the 588-kev 4.4-minute activity at 22.8 Mev was found to be 1.63±.1. This number was used to normalize the activation functions from which curves A and B of Fig. 7 were extracted.

(2) The calculated cross sections and yields of Sec. IV can be employed for the normalization of the 79-hour and 4.4-minute yield curves. It is fortunate for the purposes of normalization that the ratio of these yields at $E_{th}+1150$ kev is insensitive to the value of r chosen for Zr^{90}. At $\epsilon=1150$ kev, the calculated yield ratio (79-hour/4.4-minute)=1.52 for $r=0.8$, 1.4 for $r=0.24$, and 1.59 for $r=2.4$. Thus, the yield ratio at $\epsilon=1150$ kev is 1.5±.1 for reasonable values of r. This value of 1.5±0.1 is in good agreement with the value implied by the measured relative yields at 22.8 Mev. By using the yield curves themselves (Figs. 4 and 5), the yield ratio at $\epsilon=1150$ kev can be determined from the yield ratio at 22.8 Mev to be 1.44±.15. While the normalization obtained by either of these methods is not very precise, it is sufficiently good to allow something to be said about the distinctive features of the shapes of the cross sections. Curve C of Fig. 6 represents the cross section for the direct production of the 79-hour state and is obtained by subtracting 93% of the properly normalized 4.4-minute yield from the 79-hour yield and then extracting the cross section of this direct production activation function. To obtain curve C, the calculated yield ratio at $\epsilon=1150$ kev of 1.5 was used.

(B) Interpretation of the Relative Cross Sections

The narrowness of the 79-hour cross-section peak confirms previous observations that the total (γ,n) cross section is more sharply peaked than usual when the bombarded nucleus has its nucleons in closed-shell configurations.[43] The shape of the total formation cross section agrees well with the total (γ,n) cross section measured by Yergin and Nathans,[44] while the shape of the formation cross section of the 4.4-minute Zr^{89m} agrees reasonably well with the earlier measurement of Katz *et al.*[45]

It is interesting to note that the 4.4-minute Zr^{89m} formation cross section has a different shape from the total (γ,n) cross section. This difference in shape implies that as the energy varies, there is a change in the fraction of the (γ,n) events which lead to the 4.4-minute isomeric level. The rise in curve C of Fig. 7 above 19 Mev is probably due to the $(\gamma,2n)$ reaction of Zr^{91}. If this interpretation is correct, the $Zr^{91}(\gamma,2n)$ reaction is much more likely to produce the ground state than the 4.4-minute isomeric level in Zr^{89}. It is not unreasonable to expect such behavior since the ground-state spin of Zr^{91} is believed to be 5/2.[46]

Further analysis of the cross sections is possible only with a more complete theory of gamma-ray cascading than now exists. Since most of the gamma-ray absorption cross section at about 17 Mev is due to $E1$ absorption, the excited states of Zr^{90} are then mostly 1−. If the energy-level density of Zr^{89} is a function of spin-parity were known (or could be postulated), it should be possible to determine the relative probability of exciting each type of excited state on the basis of the foregoing discussion and calculations. If more were then known about gamma-ray cascading, experimentally determined (γ,n) cross sections for the production of pairs of isomers would provide a sensitive test of any theory dealing with energy-level densities.

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[43] R. Nathans and J. Halpern, Phys. Rev. **93**, 437 (1954).
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[45] Katz, Baker, and Montalbetti, Can. J. Phys. **31**, 250 (1953).
[46] D. H. Arroe and J. E. Mark, Phys. Rev. **76**, 873 (1949).