

## Neutron Yields from Photo-Disintegration by Gamma-Rays from Lithium\*

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The relative neutron yield of 33 elements has been studied using the gamma-ray spectrum which arises from proton bombardment of Li. The samples which were to be irradiated were placed in a block of paraffin. The neutrons were detected using four boron trifluoride proportional counters. Absolute gamma-ray and neutron intensity measurements yielded the value  $(0.055 \pm 0.012)$  barn for the  $(\gamma, n)$  cross section of copper. Rough indications were also obtained concerning the relative neutron energy distribution from the samples.

### I. INTRODUCTION

DURING recent months a number of experiments<sup>1-5</sup> have been performed to obtain information concerning the  $(\gamma, n)$  photo-disintegration processes. These experiments have been performed using either the radiation from a nuclear reaction process such as  $\text{Li}^7(p, \gamma)\text{Be}^8$  or the bremsstrahlung spectrum from a betatron. Though activation methods have been used for the detection of the disintegration by both kinds of radiation, direct neutron detection seems to have been limited to the case of disintegrations produced by the betatron.

The experiments to be described below apply the method of neutron detection to the disintegrations produced by the gamma-rays from the Li reaction. These experiments have been briefly described earlier.<sup>6</sup> The gamma-ray spectrum for the Li reaction has been examined in detail by Walker and McDaniel.<sup>7</sup> In these experiments 750-keV protons were used to bombard a thick lithium target. Under these conditions the gamma-ray spectrum consists of two lines. One of these is located at 17.6 MeV and is presumably but a few kilovolts wide. The second component is located at about 14.8 MeV and is of the order of 2 MeV in width. The intensity of the 17.6-MeV line is about twice that of the low energy component. The measurements of neutron yield which were made in the present experiment constitute a weighted average of the cross section over this spectrum and over the isotopes present in the sample. The yields of 33 samples were measured relative to the yield of copper. For copper, a measurement of the absolute value of the cross section was made.

### II. MEASUREMENT OF THE RELATIVE YIELD

The experimental arrangement which was used in the measurement of the relative neutron yield is shown in

Fig. 1. A large rectangular block of paraffin was placed immediately in front of the cyclotron target. Four enriched  $\text{BF}_3$  proportional counters, arranged in a square array, were embedded in a movable section of the paraffin. The neutron-producing sample was placed in a cavity in the paraffin located between the counters and the cyclotron target. A layer of 1 g/cm<sup>2</sup> of  $\text{B}_4\text{C}$  was placed between the sample and the counters. This absorber causes the counters to be insensitive to neutrons which are thermalized in the vicinity of the sample. Thus, variations in the slow neutron absorption cross section of the sample will not affect the slow neutron detection efficiency of the counters.

The counters were connected in pairs, front and rear, to form two counting channels. The proportional counter pulses were amplified and passed into scaling and recording circuits in two similar channels. The principal reason for the use of two counting channels was to obtain some information concerning the neutron energy spectrum of the various samples examined. It was thought that if there were radical differences between neutron spectra, this would evidence itself by a variation of the ratio of neutron counting rates in the front and rear channels.

Three lead-shielded Geiger counters were located above the cyclotron target and were used as monitors of the gamma-ray intensity.

A set of measurements on a sample consisted of consecutive observations of the counting rates in the two channels under three conditions: (a) background with no sample, (b) sample or "radiator" producing neutrons placed in the cavity, (c) standard copper radiator in cavity. All samples covered the same area, the different substances differing only in thickness. Powder samples were enclosed in thin-walled Al boxes. Their comparison measurements (a) and (c), respectively, were taken with an empty Al box, and with one containing the copper standard. In the case of heavy elements, a total of three sets of observations were taken while in the case of the light elements, five sets of observations were taken because of the lower neutron yield and consequent inaccuracy of the individual measurements. For the standard copper radiator, the ratio of the sample counting rate to the background rate was about three to one. For the samples of the heavier elements this

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<sup>1</sup> H. Wäffler and O. Hirzel, *Helv. Phys. Acta* **21**, 200 (1948).

<sup>2</sup> J. McElhinney (unpublished).

<sup>3</sup> G. Friedlander and M. L. Perlman, *Phys. Rev.* **74**, 442 (1948); **75**, 988 (1949).

<sup>4</sup> G. A. Price and D. W. Kerst, *Phys. Rev.* **77**, 806 (1950).

<sup>5</sup> L. Katz *et al.* (to be published).

<sup>6</sup> Walker, McDaniel, and Stearns, *Phys. Rev.* **79**, 242 (1950).

<sup>7</sup> R. L. Walker and B. D. McDaniel, *Phys. Rev.* **74**, 315 (1948).

ratio was frequently higher but for the lighter elements the ratio approached unity and in some cases, as will be explained below, was even less.

The calculation of the relative cross sections from the counting rates was made in the following manner. The counting rates with the sample in position were first corrected for two effects which alter the background component when the radiator is inserted. The backgrounds were then subtracted to obtain the relative number of neutrons emitted by the sample. These numbers were then corrected for the reduction, by absorption, of the gamma-ray intensity as the gamma-rays traverse the sample. The neutron yields from the samples were reduced to obtain the yield per atom. The correction for self-absorption of the gamma-rays in the radiator included a small correction for obliquity of incidence. For some of the heavy elements the self-absorption correction was as great as 30 percent.

For light elements the two effects which alter the background upon insertion of the radiator actually reduce the counting rate of the counter with radiator below that obtained with no radiator.

The first of these effects to be considered is the following. About 50 percent of the background counting rate arises from production of neutrons in the walls of the counters themselves by the incident gamma-ray flux. When the radiator is inserted, the gamma-ray flux on the counters is diminished by the absorption of the gamma-rays in the sample. This reduces the number of neutrons produced in the counter walls. In the case of light elements the neutron production may be very small and the reduction in the background may exceed the increase in the rate due to generation of neutrons in

the sample. Because the cross section for neutron production decreases more rapidly with  $Z$  than does the total cross section, the effect is larger for low values of  $Z$ .

In the case of copper the correction for this effect was about seven percent of the cross section, and was as great as 100 percent for the case of calcium. The reduction of the background due to the insertion of the radiator is obtained by multiplying the fraction of the background originating in the counters by the factor  $(1-T)$  where  $T$  is the gamma-ray transmission of the sample. The fraction of the background which arises in the counters was determined by a series of measurements which were made with the counters alternately bare and surrounded by an amount of brass equal to that of the counters. The calculations were made in such a manner as to take into account the change in the neutron sensitivity of the counters by the insertion of the additional brass. The transmission coefficients of the samples were calculated from the total gamma-ray cross section of the materials. These cross sections were based on the values measured by Walker<sup>8</sup> at 17.6 Mev. For the values of  $Z$  not directly measured by Walker, the values were determined by interpolation. The corrections to the total gamma-ray cross sections due to the presence of the 14.5-Mev line were negligible. While gamma-rays may be degraded by Compton scattering and consequently not removed from the beam, it was calculated that because of the high threshold for the  $(\gamma, n)$  reaction and the small fraction of high energy gamma-rays in the scattered radiation this effect could be neglected.

The second effect which contributes to a reduction in the counting rate on insertion of the absorber is the scattering of neutrons away from the counters. Some neutrons were generated in the vicinity of the target and could pass through the paraffin shielding to be detected in the counters. When a thick sample is placed in the beam, part of the neutrons are scattered away from the counters. To measure this effect a thick sample of carbon was placed in the beam. Since the gamma-ray threshold energy for neutron production in carbon exceeds 17.6 Mev, it was presumed that no neutrons were generated in the sample. When the carbon sample was inserted a reduction in the counting rate was observed. Much of the reduction was due to the effect first described but about 40 percent was due to neutron scattering. The determination of the magnitude of the scattering effect in carbon, used with the measured values for the neutron scattering cross section<sup>9</sup> for neutrons of about 1 Mev, permit a correction to be made for the scattering in the various samples which were used. For most samples heavier than aluminum the correction was negligible, but for the lighter elements the corrections were as great as 15 percent.

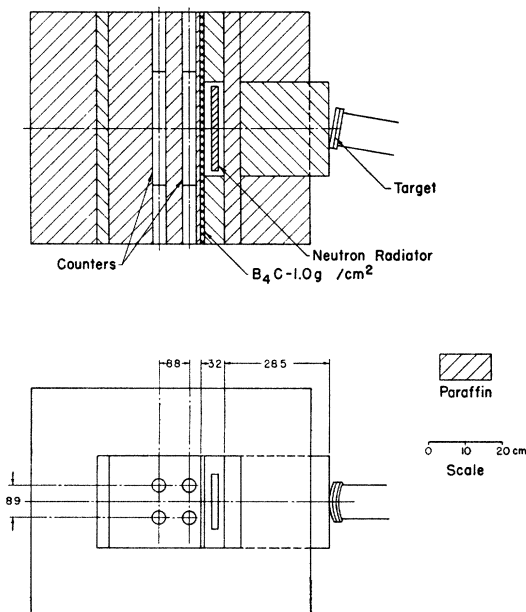


FIG. 1. Geometrical arrangement of Li target, paraffin neutron moderator,  $\text{BF}_3$  proportional counters and neutron producing sample.

<sup>8</sup> R. L. Walker, Phys. Rev. **76**, 527 (1949).

<sup>9</sup> Goldsmith, Ibsen, and Feld, *Science and Engineering of Nuclear Power, Neutron cross sections of the elements* (Addison-Wesley Press, Inc., Cambridge, 1947).

After calculation of the relative yields the results for the two channels were averaged. The results of the calculations are shown in Fig. 2. Here we have plotted as ordinate the neutron yield per atom relative to that of copper. The abscissa is  $Z$ . The smooth variation of the yield in the range of  $Z$  extending from 21 to 58 is rather striking. In this range, with the exception of Ni, no value departs from the curve shown by an amount greater than about 20 percent. Nickel is notably low, having only about one-half the value of the ordinate of the curve at this point. The yield values for  $Z$  below 20 are all low but fluctuate widely as might be expected. At high  $Z$ , Hg, Pb, and Bi seem to show a slight falling off. Uranium lies above the extrapolation of the curve, probably because of an appreciable photo-fission cross section at 17.6 Mev combined with a multiplicity of fission neutrons.

### III. NEUTRON ENERGY SPECTRUM

As was mentioned above, it was hoped that one would be able to obtain some indication of the neutron energy distribution from the various samples by comparing the counting rates of the front and rear channels. After subtraction of the background it was found that the ratio of the front to rear channel counting rates increased with increasing  $Z$  by about 35 percent. This indicates a decrease in neutron energy with increasing  $Z$ . For the most part this variation was rather systematic though a few elements deviated notably from a systematic variation. Tungsten, one of these elements, seemed to have a remarkably low energy neutron distribution.

To explore this further, measurements were made of the neutron counting rate for several samples as a function of position of the counter in the paraffin behind the radiator. This position could be varied by moving the paraffin section, which contained the counters, back and forth in the outer paraffin shell. Layers of paraffin were added between the sample and the counters to keep the spaces filled with paraffin at all times. The results of the measurements are shown in Fig. 3. Here we have plotted the ratio of the corrected counting rate of the sample to that for copper. The abscissa is the distance  $D$  between the face of the cavity in the paraffin and the center of the counters. We see that the curve for tungsten falls off much faster than that of Pb. For a comparison standard the neutrons from Ra- $\alpha$ -Be and Po- $\alpha$ -Be were also examined. The counting rates for these spectra are also plotted on the curve relative to the Cu counting rate. Both of the natural sources seem to indicate the presence of appreciably higher energy neutrons than are present either in the spectrum of copper, Pb or W. It is not possible to obtain from such an experiment as this the actual neutron spectrum from the elements examined but the curves serve to indicate the gross differences between the spectra. Since the average energy of the Po- $\alpha$ -Be source is about 4 Mev, and the low energy component of the Ra- $\alpha$ -Be is

probably less<sup>10</sup> than 1 Mev, one may guess the average energy of the neutrons from Cu to be about 1 Mev.

### IV. DETERMINATION OF Cu CROSS SECTION BY MEASUREMENT OF ABSOLUTE GAMMA-RAY AND NEUTRON INTENSITY

The determination of the absolute yield of neutrons from the radiator was made by a comparison between the neutron counting rate from the radiator and that from a standard Ra- $\alpha$ -Be source with the counters located in their standard position. When the gamma-rays from the cyclotron target fall on the radiator, the illumination of the radiator is not uniform. Furthermore, the sensitivity of detection of the neutrons which originate from various parts of the radiator is not constant. To take these effects into account in making the calibration using the Ra- $\alpha$ -Be source, observations were made in the following manner. An aluminum plate,

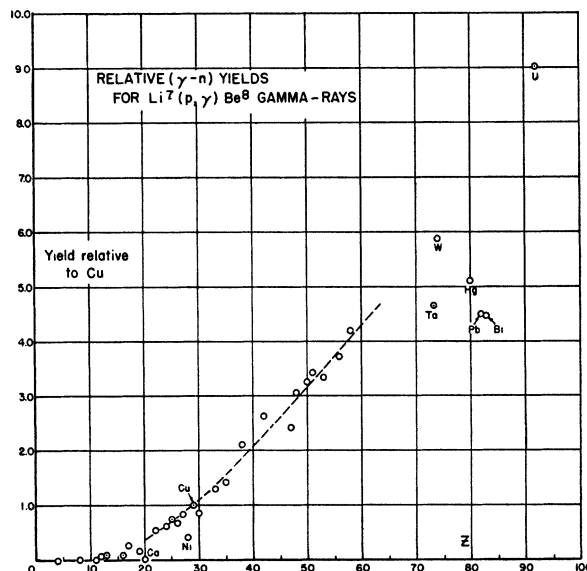


FIG. 2. Neutron yield per atom relative to that from copper.

the size of the radiator, was subdivided into 25 smaller areas. In the center of each area was drilled a hole to receive the RaBe neutron source. The plate was placed in the radiator position and the counting rate was observed for the source placed in each of the 25 source positions in the plate. The observed rates were then weighted by a factor proportional to the area represented by each location and proportional to the relative intensity of illumination of this area when irradiated by gamma-rays from the cyclotron. The intensity of illumination from the cyclotron was assumed to follow the inverse square law. The neutron detection efficiencies were found to be about one percent and one-half percent for the front and rear counter pairs, respectively.

The determination of the gamma-ray intensity was

<sup>10</sup> H. L. Anderson, Nuclear Science Series, Preliminary Report No. 3, National Research Council.

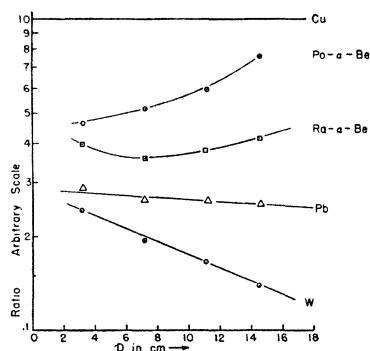


FIG. 3. Relative spatial distribution of neutron density in paraffin. Counting rate compared to that of standard Cu sample as function of distance  $D$  from center of counter to face of cavity in the paraffin which contains sample. Ratios have been multiplied by arbitrary factors and plotted on a logarithmic scale.

based upon a direct measurement of the number of Compton and pair processes produced in a thin radiator by the gamma-ray flux. This method was used earlier by Hough.<sup>11</sup> As is indicated in Fig. 4, a thin-walled Victoreen Geiger counter was placed in the gap of a large magnet so the field lines were directed along the axis of the counter. The air gap of the magnet was heavily shielded by lead. In the direction of the cyclotron target there was an opening in the shielding which permitted the gamma-rays from all parts of the target to strike a chosen region,  $1.5 \times 1$  cm, on the counter. Thin radiators of Pb, Cu, and Al were placed immediately in front of the counter. The counting rate of the thin-walled counter was measured as a function of the thickness and material of the radiator. The gap height of the magnet was 15 cm and the pole diameter was 35 cm. The target to counter distance was 105 cm.

For sufficiently thin radiators it is expected that either the Compton process or the pair process will always give rise to a count in the counter. The presence of the magnetic field aids in this respect by increasing the probability of counting low energy electrons which are scattered through large angles. Because of the resolving time of the counters, one, and only one, count is obtained for each process. Thus the incident gamma-ray flux can be determined from a knowledge of the counting rate, the geometry and the total cross section of the material of the radiator for the gamma-ray spectrum being used. The principal function of the magnetic field is to lower the background by preventing electrons which are produced in the walls and collimator from reaching the counter. To decrease the number of electrons scattered from the walls of the chamber, the walls were covered by  $\frac{1}{4}$  in. of wood. Because of the large increase in the total cross section at low energies by the Compton and photoelectric processes, it is necessary to determine that there is not a sufficient amount of degraded radiation present to cause difficulty in the measurements. To check this

<sup>11</sup> P. V. C. Hough (unpublished).

possibility a lead absorber was placed immediately behind the collimator. Its absorption coefficient was found to be that expected for the high energy radiation present in the spectrum. A further check on this point is the good agreement which was found between the results for the radiators of the three different elements. The background counting rate in the counter with no radiator present was quite low and was insensitive to the magnetic field strength. The value of field strength which was used was 3000 gauss.

The results of the gamma-ray intensity measurements are shown in Fig. 5. The ordinates plotted are the ratios of counting rate of the Victoreen counter to the monitor rate. Plotted on the abscissa is the grams of material contained in the radiator. It will be noticed that the increase in counting rate with radiator weight is quite linear to rather large values. From the slopes of the curves and from the gamma-ray cross-section values one can obtain the gamma-ray flux corresponding to a given monitor rate. The cross-section values which were used were taken from the measurements by Walker. A small correction was made in the case of lead for the fact that Walker's measurements<sup>8</sup> were taken at 17.6 Mev whereas the gamma-ray spectrum used here contained some intensity at 14.8 Mev. For copper and aluminum no correction was necessary. The results of the intensity measurements for the Pb, Cu, and Al agreed to within four percent of the average of the three measurements.

This method of measuring gamma-ray intensities was checked at an earlier time by another method which utilized the calculated efficiency of the gamma-ray pair spectrometer of Walker and McDaniel.<sup>7</sup> The results of the measurements by the two methods agreed to five percent. The calculated source strength was about  $1 \times 10^7$  gamma-rays per second.

Price and Kerst<sup>4</sup> examined the angular distribution of neutrons from lead and iron and found it to be isotropic. In the present experiment it was assumed that

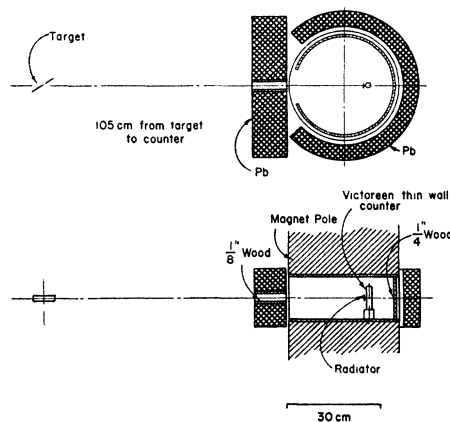


FIG. 4. Arrangement for the measurement of absolute gamma-ray intensity. Gamma-rays from target produce Compton recoil and electron pairs in the radiator in front of counter. Magnetic field serves to reduce counter background and improve detection efficiency for low energy electrons.

all elements examined had an isotropic neutron distribution. Devons and Hine<sup>12</sup> have shown that the gamma-ray production in the Li reaction deviates slightly from isotropy for protons whose incident energy is that of the resonance energy. Though our proton energy was 750 kev, since a thick target was used, nearly all of the gamma-ray intensity arises from resonance protons. The two sets of apparatus for the neutron measurement and the gamma-ray measurement were located at slightly different angles with respect to the direction of the incident proton beam. This required a correction of three percent in the absolute cross-section measurement because of the asymmetry in gamma-ray production.

The value obtained for the Cu cross section for the  $(\gamma, n)$  process is 0.055 barn. This is the average of the two values arising from the neutron counting rates of the front and rear counting channels. It is also the average for both isotopes in the sample and both gamma-ray components present in the spectrum. Because of the uncertainty resulting from the difference between the neutron spectrum of the radiator and the calibrating neutrons source, it is difficult to estimate the over-all accuracy of the measurements. Our estimate of the probable error is 0.012 barn.

#### V. DISCUSSION

It is difficult to make a reliable comparison between the results obtained in this experiment and those of other experiments because of the widely different conditions. Wäffler and Hirzel<sup>1</sup> obtained their data using the Li gamma-ray spectrum but used activation methods for detecting the neutron emission process. Price and Kerst<sup>4</sup> measured the neutrons directly but used the broad bremsstrahlung spectrum. Friedlander and Perlman,<sup>3</sup> and McElhinney<sup>2</sup> used activation methods with a bremsstrahlung spectrum.

In order to make a comparison with other experiments our results are replotted in Fig. 6, on a logarithmic cross-section scale. Our points are indicated by the open circles. The heavy black dots indicate the values obtained by Wäffler and Hirzel while the dashed curve is that obtained by Price and Kerst using the 22-Mev bremsstrahlung spectrum. For our data and those of Wäffler and Hirzel, the absolute cross-section values are plotted. In the latter case it was necessary to average the isotopic cross sections to permit a comparison. The curve of Price and Kerst has been normalized arbitrarily to match our data at  $Z$  equal to 29. The indicated errors in our data represent the standard deviation of the average of the various runs on the individual samples. It is likely that the systematic errors are equally large or larger, in view of the high background and large corrections which are applied. The values given for  $Z$  of 20 or less are quite inaccurate but should serve as a rough measure of the cross sections.

<sup>12</sup> S. Devons and M. G. N. Hine, Proc. Roy. Soc. A199, 56-73 (1949).

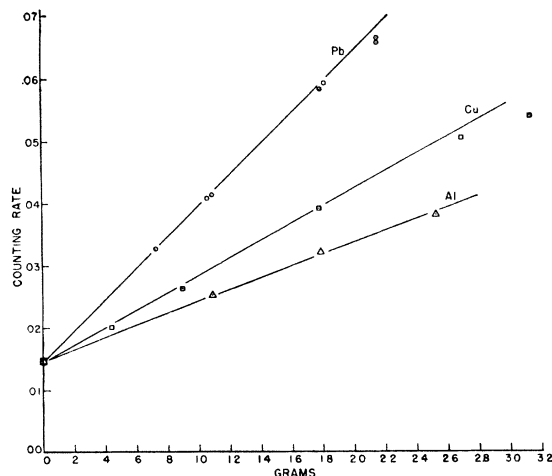


FIG. 5. Counting rate compared to monitor rate for Victoreen counter used in the measurement of absolute gamma-ray intensity. Counting rate is plotted as a function of radiator weight. Area of radiator was 1.5 cm<sup>2</sup>.

Though many of the values of Wäffler and Hirzel are in fair agreement with ours, there are several values which differ widely. For  $Z$  greater than 20 the values obtained in our experiment lie rather smoothly along a curve, but the values of Wäffler and Hirzel fluctuate rather severely. This disagreement in the relative values of the cross section may perhaps be explained on the basis that the quantities measured in the two experiments are different. By necessity the activation method chooses particular isotopes of certain elements whereas the direct neutron measurement is an average over all isotopes for each of the available samples. It may be argued further that the neutron experiments give heavy weight to possible  $(\gamma, 2n)$  reactions. However, the measurements of Perlman and Friedlander at 50 Mev for  $F^{19}$ ,  $P^{31}$ , and  $Cu^{63}$  indicate that the  $(\gamma, 2n)$  process is at most 10 percent of the  $(\gamma, n)$  cross section for the light elements. The  $(\gamma, 2n)$  thresholds for most of the elements examined probably exceed the 17.6-Mev gamma-ray energy. For those cases in which the  $(\gamma, 2n)$  reaction is possible one would expect the yield to be small because of the nearness to the threshold. One would expect that the absolute cross section measured by activation would be equal to, or smaller than, the cross section computed from the neutron yield assuming the emission of a single neutron. Of the elements measured both by Wäffler and Hirzel and by us, Cu is outstanding in that their value is 2.5 times greater than ours. While there are several other elements for which their values are notably high as compared with a smooth curve through our points, in these cases we have no measurements on elements of the same  $Z$ .

The agreement between our data and that of Price and Kerst seems to be much better. Though they also obtained data using the bremsstrahlung from 18-Mev electrons as well as those of 22 Mev, it was felt that it was better to compare our data with the 22-Mev data

rather than with either the 18-Mev data or the difference between the two. For the 18-Mev bremsstrahlung spectrum there are very few gamma-rays of energy as great as 17.6 Mev. With this spectrum one would expect the yield to depend strongly on the threshold for the reaction rather than on the cross section near 17 Mev, since the maximum in the cross-section curve probably occurs near or above 17 Mev for most of the elements. The difference spectrum is but little better in this respect and has the additional disadvantage of being very sensitive to the calibration of the gamma-ray

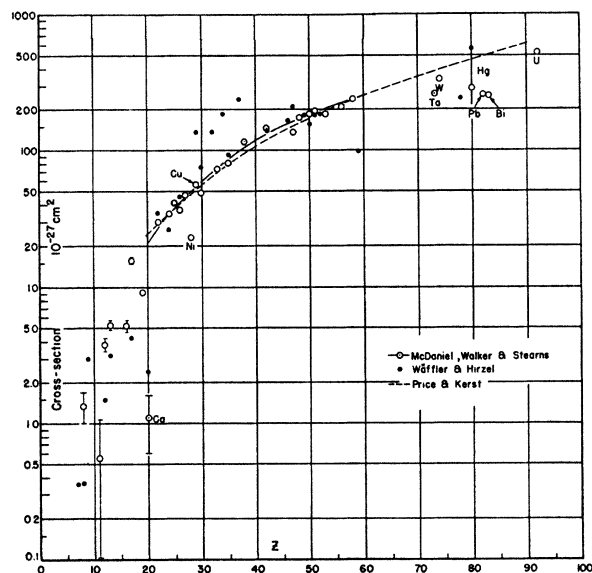


FIG. 6. Neutron yield plotted as a function of  $Z$ . The yield is plotted as  $(\gamma, n)$  cross section per atom. Data of Price and Kerst and McDaniel, Walker, and Stearns are plotted assuming no multiplicity of neutrons in the emission process. Errors shown on circled points are based on calculation of standard deviations. Systematic errors may be appreciably greater.

intensity in the two cases. The comparison with the 22-Mev data was not extended below  $Z = 20$  because the fluctuating and increasing values of the threshold for neutron production would make the comparison of little significance.

Katz and co-workers have measured the  $(\gamma, n)$  activation cross section of  $\text{Cu}^{63}$  and  $\text{Cu}^{65}$  using the bremsstrahlung spectrum. Assuming the shape of the spectrum they have obtained a differential cross-section curve as a function of energy. The relative values which they obtain at 17.6 Mev for  $\text{Cu}^{63}$  and  $\text{Cu}^{65}$  are in wide disagreement with those obtained by Wäffler and Hirzel. They find  $\sigma^{65}/\sigma^{63} \approx 0.2$ . Wäffler and Hirzel obtain the value 1.5 for the same ratio. If one takes Katz's absolute values for the differential cross section and averages

over the two isotopes and over the Li spectrum one obtains the value 0.056 barn for the cross section, in agreement with our results.

By the Teller-Goldhaber<sup>13</sup> theory, one expects the  $(\gamma, n)$  cross section to show a strong peak as a function of energy. Both the Goldhaber-Teller theory and the Levinger-Bethe theory<sup>14</sup> predict that the mean energy of the photon absorption spectrum should decrease with increasing  $Z$ . However, neither theory indicates what one would expect for the variation of the cross section as a function of  $Z$  while using a monochromatic line at 17.6 Mev.

Other experiments<sup>15-17</sup> have shown the expected decrease of resonance energy with increasing  $Z$ . One feature in our results may also indicate this. It will be observed that above  $Z = 74$ , our data indicate a decrease in the yield whereas the data of Price and Kerst indicate an increase with increasing  $Z$ . It seems possible that with increasing  $Z$  the resonance energy has fallen below the 17.6-Mev line giving rise to a decrease in the yield at that energy, whereas the yield averaged over the betatron spectrum continues to increase in about the same manner as for lighter nuclei, more-or-less independently of the location of the resonance. Because of the observed low value of the resonance energy<sup>5, 16</sup> in Ta, one is not surprised to find that Ta also yields a low cross-section value for the Li spectrum.

It would seem to be very desirable to repeat the measurements using the high energy gamma-ray from the reaction  $\text{H}^3(p, \gamma)\text{He}^4$ . Such an experiment should give additional information concerning the shape of the  $(\gamma, n)$  cross-section curve as a function of energy and  $Z$ .

The cross sections of Ca and Ni are notable in that they deviate so far from the smooth curve which can be drawn through the other points. These deviations have also been observed in the other experiments.<sup>3, 4</sup> Though these fluctuations may be, in a sense, accidental, one may hope that they will be explained by a correct theoretical interpretation. Measurements of the  $(\gamma, p)$  and  $(\gamma, 2n)$  cross sections would perhaps shed some light on this question.

We wish to thank Dr. Bruce Dayton for his assistance in the operation of the cyclotron. We are indebted to the Argonne National Laboratory for the calibration of the Ra- $\alpha$ -Be neutron source strength and to Dr. L. Katz for the communication of his results prior to publication.

<sup>13</sup> M. Goldhaber and E. Teller, Phys. Rev. **74**, 1046 (1948).

<sup>14</sup> J. S. Levinger and H. A. Bethe, Phys. Rev. **78**, 115 (1950).

<sup>15</sup> G. C. Baldwin and E. C. Klaiber, Phys. Rev. **73**, 1156 (1948).

<sup>16</sup> McElhinney, Hanson, Becker, Duffield, and Diven, Phys. Rev. **75**, 542 (1949).

<sup>17</sup> K. Strauch, Phys. Rev. **78**, 84 (1950); **79**, 241 (1950).