Systematics of Photoneutron Reactions*

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Properties of the giant dipole resonances for (γ,n) reactions have been measured for 14 singly-isotopic elements distributed throughout the range of z values. Systematics are reported for the behavior of the integrated cross sections, the energies at which the dipole resonances attain a maximum cross section, the values of the cross sections at these energies, and the widths of the resonances. Anomalously narrow widths are reported for elements with neutron numbers in the vicinity of the magic numbers.

INTRODUCTION

EARLY experimental studies[1–5] of the absorption by nuclei of photons with energies in the range from 5 Mev upwards have shown that the absorption process exhibits a giant resonance in the energy region about 20 Mev for all elements investigated, and a considerable amount of recent work has substantiated this conclusion.[6] Theoretical investigations of photonuclear processes have established that the photon absorption is predominantly dipole in nature.[7,8] One of the fundamental predictions of the theories arises from the application and extension of the sum rules for dipole absorption to absorption by nuclei, and in fact a measurement of the total integrated cross section for dipole absorption could yield a value for the percent of exchange in the neutron-proton interaction.

Experimental techniques, however, have not yet reached the state where such comparison with theory can be made with profit. Difficulties arise from the bremsstrahlung nature of the photons used, from the lack of certainty that one has measured all partial reactions resulting from the absorption process, and from lack of knowledge of any tails on the absorption curve above the giant resonance and what fraction of any tail arises from dipole absorption. On the other hand, a detailed study of the parameters of the giant dipole resonance itself and of the dependence of these parameters on atomic number can yield direct information concerning nuclear structure and is less exacting in experimental requirements, especially if the assumption is made that the excitation function for a photoneutron reaction is identical except in magnitude with that for total photon absorption over the giant dipole resonance.

With this in mind, we have made a study of the photoneutron cross sections of 14 singly-isotopic elements from threshold to 24 Mev using betatron bremsstrahlung and direct neutron detection. The practical advantages of direct neutron detection over residual activity measurements in obtaining precise yield curves are so great as to outweigh any ambiguities arising from the onset of $(\gamma,2n)$ or (γ,pn) reactions. During the course of this study there appeared a similar work by Montalbetti, Katz, and Goldenberg[9] in which many of the elements herein investigated were measured. The apparatus used for the work of this paper had a signal-to-background ratio an order of magnitude larger than that used by Montalbetti *et al.* and consequently comparatively thinner samples could be employed. Preliminary results of this work have recently been published.[10–12]

APPARATUS AND PROCEDURE

The general arrangement of the apparatus is shown in Fig. 1. The details of construction as well as the significant performance data have been presented in previous publications.[13,14] The only modification for the present study was the use of four enriched BF_3 counters imbedded symmetrically at 13.5 cm from the beam axis to enhance the yields near threshold and reduce the exposure time for a given element to reasonable limits. The samples, whose neutron yields are measured as the betatron is run at successively increasing energy in half-million volt steps from threshold to 25 Mev, are situated in the center of the paraffin house at 296-cm distance from the betatron target. At this distance the collimated betatron beam has a diameter of 1.03 cm and an intensity of 3.9 r/min at 20-Mev betatron energy. Some samples were in powder form and were packed into aluminum cylinders with walls 0.030 in. thick and end caps 0.007 in. thick. Identical blank cylinders permitted background determinations for these elements.

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[6] We shall refer to individual papers that pertain to the results of this paper in subsequent sections. A relatively complete bibliography of papers on photonuclear reactions is given in the paper by L. Eyges, Phys. Rev. **86**, 325 (1952).
[7] M. Goldhaber and E. Teller, Phys. Rev. **74**, 1046 (1948).
[8] J. S. Levinger and H. A. Bethe, Phys. Rev. **78**, 115 (1950).
[9] Montalbetti, Katz, and Goldenberg, Phys. Rev. **91**, 659 (1953).
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[13] Halpern, Mann, and Nathans, Rev. Sci. Instr. **23**, 678 (1952).
[14] Sher, Halpern, and Mann, Phys. Rev. **84**, 387 (1951).

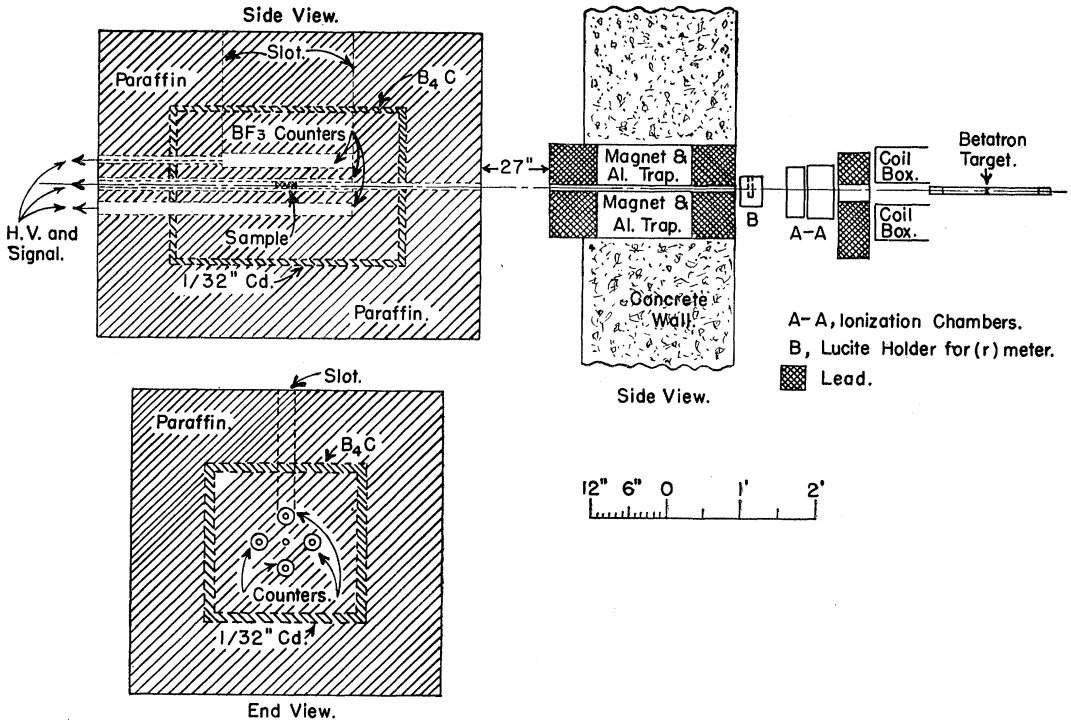

FIG. 1. General arrangement of the apparatus.

With the paraffin house modified to contain four BF_3 counters, it was necessary to establish the dependence of counting efficiency on the energy of the neutrons leaving the sample for each of the counters. Details of this calibration have also been presented previously.[13] Within the errors involved in the calibration, the efficiency of neutron detection was in all cases independent of neutron energy. Because of variations in the BF_3 counters themselves, the absolute efficiency of the four counting channels varied somewhat as determined by a standard radium-beryllium source inserted in the paraffin house at the sample position. The efficiencies so determined were 0.611 percent, 0.553 percent, 0.447 percent, and 0.591 percent, respectively. The four channels were operated from a single high-voltage supply, while their outputs were fed into two amplifying channels, each taking two counters. Data from the two channels were independently recorded and examined for consistency.

Samples were not of sufficient thickness to warrant correction for absorption of the x-ray beam in passing through them, as was demonstrated by runs in which a given element was irradiated using two samples identical in all respects except thickness. In all cases, the yields were identical when normalized to the same thickness. A more serious problem that could arise from thick samples is the possibility of x-ray scattering into the paraffin house producing a (γ,n) reaction in the natural deuterium which is present to three parts in twenty thousand in the hydrogen of the paraffin. Absence of any such difficulty was demonstrated by equality of background with and without samples at energies below the sample thresholds.

Background arose from two sources, that of cosmic-ray origin and that arising from photoneutrons produced in the betatron target and elsewhere leaking through the paraffin house shielding. Each component of the background was separately determined so that for runs near threshold of some elements, where the background is primarily of cosmic-ray origin, corrections could be made for difference in exposure times. Figure 2 shows the values of the two background components as determined from summing the counts for the four channels.

Procedure on a run for a given element consisted in determining neutron yields at various betatron energies for a given number of roentgens of exposure as measured by a 100r Victoreen thimble imbedded in a Lucite cylinder 8 cm in diameter located 3 ft from the betatron target. Each yield was taken to a statistical accuracy of 1 percent except the first three points near threshold, where the accuracy was always better than 3 percent. The entire run was repeated at least once, and internal consistency was within the statistical error. After each run, the threshold and yield at 14.5 Mev for a standard bismuth sample were measured to insure stability of the energy scale. All such determinations indicated no energy shift of the betatron greater than 0.1 Mev. Details of the energy control and calibration have appeared in a previous publication.[14] Excitation func-

tions were constructed from the yield data using the bremsstrahlung distributions as calculated at 1-Mev intervals and modified for absorption in the doughnut wall, the ionization chamber, and the Lucite block.[15]

RESULTS

Figure 3 shows the results of a typical yield curve for a sample of 1.81 g/cm² of rhodium, with the background drawn in for comparison. For economy of figures, the yield curves for the remaining elements are not presented. Furthermore, intercomparison of yield curves from different laboratories is sometimes misleading inasmuch as such curves are the peculiar property of the special arrangement of monitoring apparatus in the x-ray beam and the modification of the bremsstrahlung distribution thereby produced. In constructing excitation functions from yields, this effect is of course taken into account. Cross-section data are presented in Fig. 4 for 12 of 14 elements investigated. The curves for bismuth and tantalum have already been published.[10] Latest results of the Saskatchewan group[9] are drawn for comparison.

The parameters of the giant resonances as taken from these figures have been presented in tabular and graphical form in a preliminary account of this work.[12] Also included were the yields at 22 Mev for comparison with the measurements of Price and Kerst.[16]

In Table I, which presents the data of this paper in more complete form, the absolute values of the cross sections and yields have been modified somewhat from

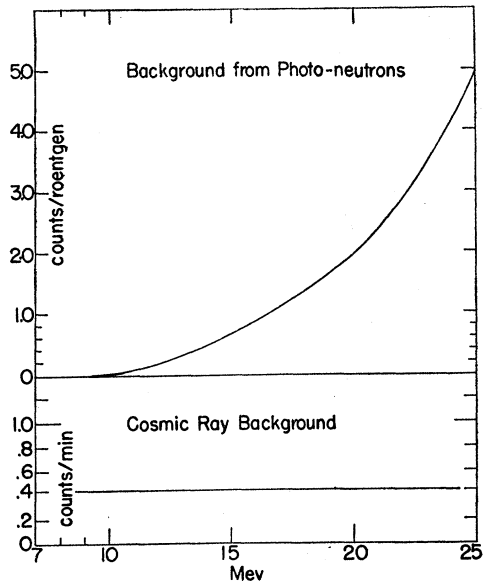

FIG. 2. The upper curve shows the background signal from photoneutrons produced in and around the betatron as a function of betatron energy. The lower curve shows the background component from cosmic rays.

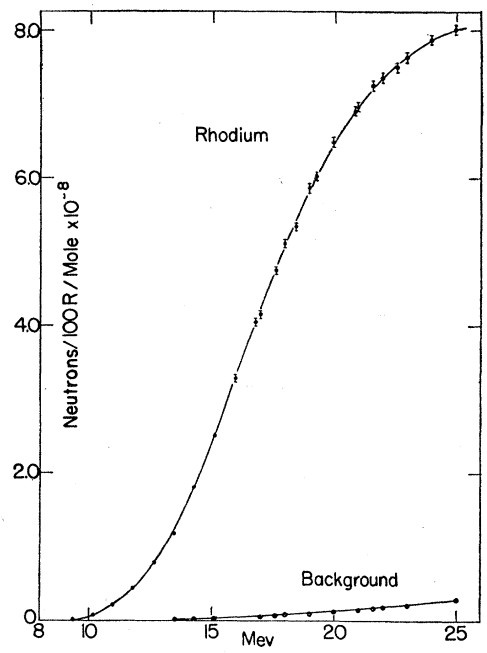

FIG. 3. Neutron yield curve for rhodium. The uncertainty on each point represents statistical error in the evaluation of the yield at each energy. Shown also is the background for comparison.

those given in reference 12. The 22-Mev yields herein given are 10 percent higher to conform to the monitoring arrangement of Price and Kerst; the absolute cross sections are 10 percent lower as a result of improved measurements of the beam area. In the case of bismuth and tantalum, however, absolute values are 40 percent and 8 percent higher, respectively, because of a previous calculational error.

McDaniel, Walker, and Stearns,[17] using the $Li^7(p,\gamma)B^8$ reaction as a radiation source and boron trifluoride proportional counters for neutron detection, have measured absolute cross sections at 17 Mev for some of the elements listed. Corresponding values from our data are tabulated for a rather stringent comparison, as will be discussed in detail below.

DISCUSSION

The parameters listed in Table I which are of special interest in a study of systematic trends in photoneutron reactions are the integrated cross section, the energy at which the dipole absorption resonance attains a maximum cross section, the value of the cross section at this energy, and the narrowness of the resonance as designated by the width in Mev at half-maximum. Each of these is separately considered below.

Integrated Cross Section

The sum rules for dipole absorption as applied to nuclei by Levinger and Bethe[8] give for the integrated

[15] Johns, Katz, Douglas, and Haslam, Phys. Rev. **80**, 1062 (1950).

[16] G. A. Price and D. W. Kerst, Phys. Rev. **77**, 806 (1950).

[17] McDaniel, Walker, and Stearns, Phys. Rev. **80**, 807 (1950).

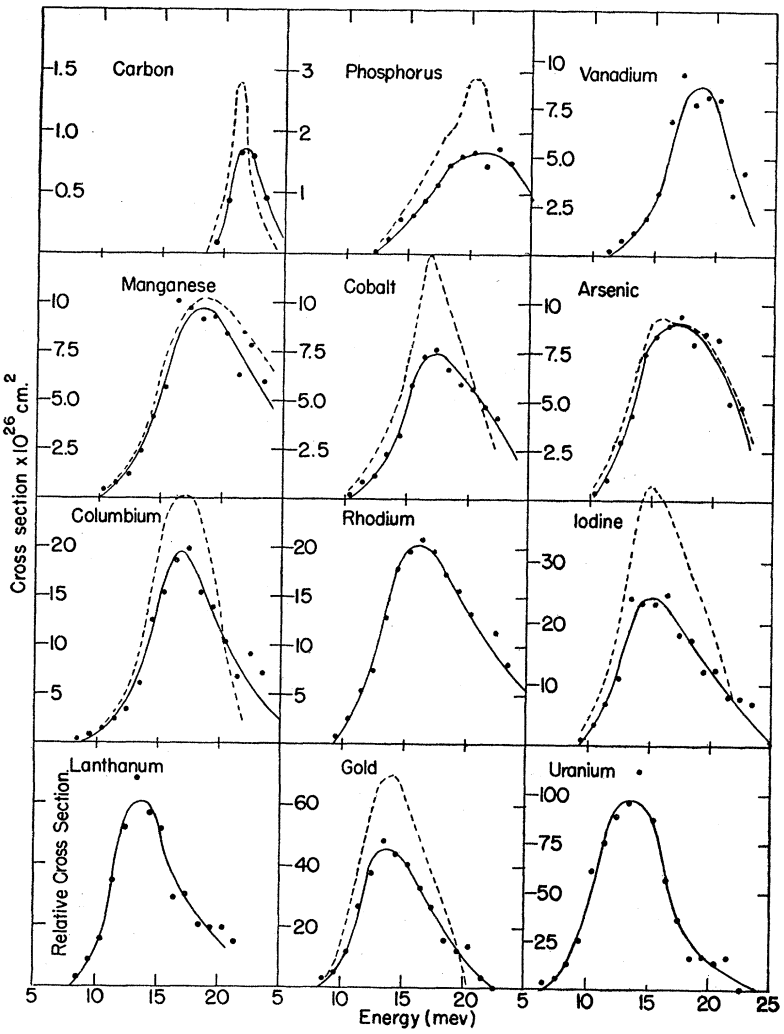

FIG. 4. Excitation functions for 12 of the 14 elements here considered. The dashed lines show the results of Montalbetti *et al.*

cross section for photon absorption:

$$\int_0^\infty \sigma dE = 0.06\frac{NZ}{A}(1+0.8x),$$

where x is the fraction of attractive exchange force in the neutron-proton interaction. Here σ is the sum of the partial cross sections for all possible reactions involving the dipole absorption of a photon. In principle, an experimental determination of the integrated cross section should permit a value of x to be obtained from the above equation. It must be noted, however, that

TABLE I. Summary of data on photoneutron reactions.

Element	Target thickness g/cm²	Thresholds (γ,n) Mev	Thresholds $(\gamma,2n)$ Mev	Thresholds (γ,pn) Mev	E_{σ_m} Mev	Half-width Mev	σ_m ×10²⁶ cm²	$\int_0^{25\text{ Mev}}\sigma dE$ Mev-barns	22-Mev yield/mole-r ×10⁻⁶ (this paper)	22-Mev yield/mole-r ×10⁻⁶ (Price and Kerst)	$\sigma_{17\text{ Mev}}$ ×10²⁶ cm² (this paper)	σ ×10²⁶ cm² (McDaniel)
$_6C^{12}$	12.80	18.70	32.60	27.41	22.0	3.0	0.86	0.027	0.028			
$_{15}P^{31}$	4.41	12.05	24.50	17.90	21.5	10.2	1.67	1.61	0.389	0.33		
$_{23}V^{51}$	2.68	11.15	19.88	18.80	18.7	5.8	8.69	0.56	2.26	1.6	6.8	3.3
$_{25}Mn^{55}$	1.57	10.00	19.40	17.72	18.4	8.8	9.69	0.88	3.15	1.9	9.0	4.0
$_{27}Co^{59}$	2.59	10.25	19.67	17.48	17.3	8.4	7.45	0.63	2.51	2.3	7.4	4.8
$_{33}As^{75}$	1.34	10.10	17.42[a]	16.82[a]	17.3	9.0	9.03	0.80	3.73	3.9	8.5	7.0
$_{41}Cb^{93}$	1.36	8.70	17.88[a]	14.08[a]	17.0	6.8	19.5	1.46	6.67	5.8		
$_{45}Rh^{103}$	1.81	9.35	17.47[a]	15.41[a]	16.5	8.9	20.5	1.94	9.00			
$_{53}I^{127}$	1.23	9.10	16.72	15.29	15.5	8.3	24.3	2.04	11.2	11.0	21.0	18.0
$_{57}La^{139}$	...	8.80	14.25[a]	14.90[a]	13.8	5.7	...	...	...	...	...	...
$_{73}Ta^{181}$	0.468	7.55	13.84[a]	13.47[a]	15.1	7.9	39.7	3.43	21.0	19.0	34.0	25.0
$_{79}Au^{197}$	0.455	7.90	13.71[a]	12.94[a]	13.9	6.9	45.6	3.19	22.4	19.0		
$_{83}Bi^{209}$	0.434	7.40	13.30[a]	12.68[a]	13.2	4.1	63.0	4.08	25.0	25.0	20.0	24.0
$_{92}U^{238}$	0.548	5.97	12.18[a]	13.34[a]	13.8	6.6	98.0	7.15	57.5	51.0	48.0	52.0

[a] Computed from mass formula.

the computed value of x is very sensitive to experimental errors in $\int\sigma dE$. For instance, with $x=0.5$, a 10 percent error in $\int\sigma dE$ leads to a 35 percent error in x. Uncertainties in $\int\sigma dE$ arise from several causes: errors in the measurement of the excitation function for a given partial reaction, neglect of some of the partial reactions, or failure to include contributions of all energy photons to a given partial cross section. For the case of the (γ,n) partial cross section where neutrons are counted, an additional error concerns lack of knowledge of the neutron multiplicity arising mainly from $(\gamma,2n)$ reactions.

Figure 5 shows the systematics of our total neutron emission cross-section measurements integrated to 25 Mev. Here $\int\sigma dE/(NZ/A)$ is plotted against A. Lines for $x=1$ and $x=0$ are shown. For elements with high Z where partial reactions involving emission of charged particles can safely be neglected, the largest contributions to the integrated cross section are those from the (γ,n) and $(\gamma,2n)$ reactions. For these elements the neutron multiplicity might be expected to decrease the values plotted by 20 percent whereas the results of Jones and Terwilliger[18] would indicate the values should be increased by the same amount because of neglect of contributions to the integrated cross sections from photons above 25 Mev. Furthermore, the magnitude of the (γ,γ) process can contribute a like amount to the integrated cross section.[19] Although these uncertainties preclude a reliable determination of x from the presently available photonuclear data, nevertheless it should be noted that the experimental integrated cross

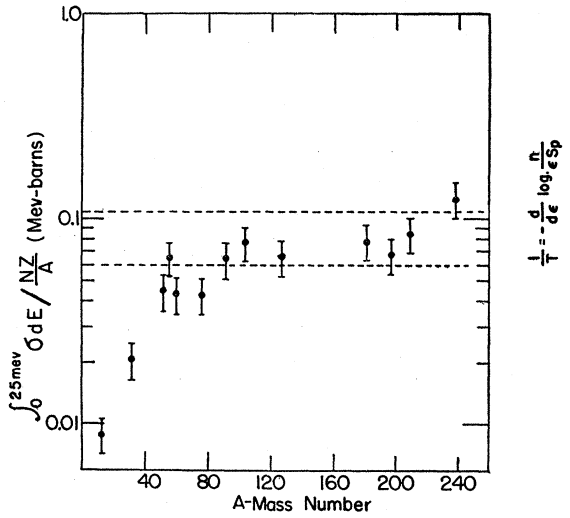

FIG. 5. Total neutron emission cross sections integrated to 25 Mev divided by NZ/A, and plotted against mass number. The straight lines indicate the results to be expected from calculation of the total integrated cross sections for dipole absorption from the theory of Levinger and Bethe for zero and 100 percent exchange force.

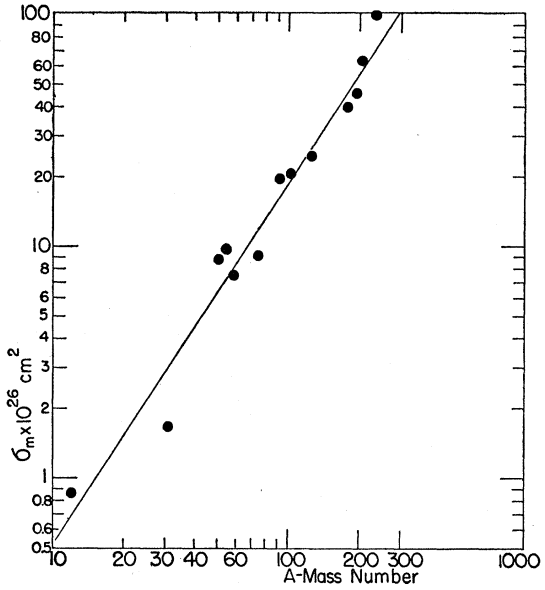

FIG. 6. Log-log plot of the maximum cross section *vs* A. The best straight line through the points gives a dependence on mass number of $A^{\frac{5}{3}}$.

sections for the heavy elements are consistent with the predictions of the sum rules.

Uranium, with a total neutron integrated cross section of 7.1 Mev-barns is of course a special case because of the added neutron multiplicity resulting from photofission. Duffield and Huizenga[20] have reported separate measurements of the (γ,n) and $(\gamma$, fission) integrated cross sections with values of 2.6 Mev-barns and 1.2 Mev-barns, respectively. With these values and the assumption that the number of neutrons emitted per fission is 2.5, we compute the $(\gamma,2n)$ contribution to be 0.75 Mev-barns, and thus the total photon absorption $\int_0^{25\text{ Mev}}\sigma dE$ to be 4.55 Mev-barns in excellent agreement with the sum rules for 50 percent charge exchange.

For light elements, neutron emission represents only a small contribution to the total photon absorption cross sections, and for $A<75$ the points of Fig. 5 fall below the line $x=0$ as might be expected.

There are still insufficient data, however, to explain the low integrated cross sections for some of these elements as exemplified by the data for carbon. The (γ,n) integrated cross section of 0.027 Mev-barns plus that for (γ,p) of 0.063 Mev-barns[21] together represent only 50 percent of the sum rule predictions for $x=0$. Should the (γ,p) excitation function exhibit as striking a tail as the (γ,n) results of Jones and Terwilliger[18] indicate, then no discrepancies exist.

We have not as yet discussed the uncertainties in the integrated cross sections arising from possible errors in our measurements. These can arise from systematic errors which may occur in determining absolute yield

[18] L. W. Jones and K. M. Terwilliger, Phys. Rev. **91**, 699 (1953).

[19] Mary Beth Stearns, Phys. Rev. **87**, 706 (1950).

[20] R. B. Duffield and J. R. Huizenga, Phys. Rev. **89**, 1042 (1953).

[21] J. Halpern and A. K. Mann, Phys. Rev. **83**, 370 (1951).

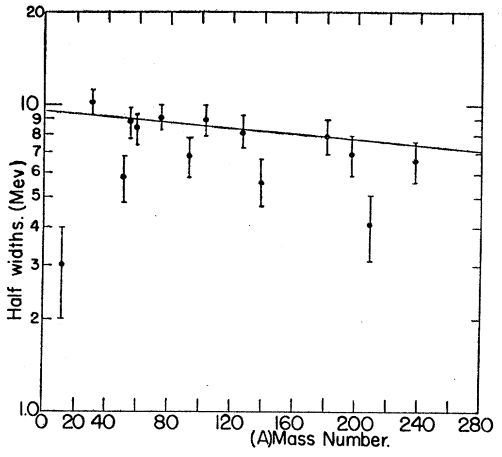

FIG. 7. Half-widths of the giant resonances as a function of mass number.

values and from the conversion of bremsstrahlung yields to cross sections at a given energy. The most significant systematic error in the determination of absolute yields arises from calibration of the efficiency of the detection system using a standard radium-beryllium source whose neutron output is known to perhaps 15 percent. All other errors should be negligibly small. The yield measured at a given maximum bremsstrahlung energy is, however, the result of contributions from photons of all energies between the threshold of the reaction and that maximum bremsstrahlung energy. To calculate the cross section at the given photon energy requires that the contributions from all photons, except these in a small energy region about the given energy, be subtracted from the measured yield. The (approximate) $1/E$ shape of the photon number spectrum in conjunction with the peaked nature of the cross section *vs* energy curve makes the sum of those contributions to be substracted a large fraction of the measured yield. For instance, in bismuth, the measured yield at 17-Mev maximum bremsstrahlung energy is 79.7 counts per r. The yield due to photons between 7.4 Mev (the threshold) and 16 Mev is 72.5 counts per r. Although the error in these counting rates is only 1 percent, errors of 25 percent in the value of a cross section at a given energy are not unlikely. From these considerations, our 17-Mev cross-sections values are in satisfactory agreement with the results of McDaniel, Walker, and Stearns.[17]

The magnitude of the possible error in the integrated cross section should be smaller than that in a cross-section value at a given energy. The reason for this is that the calculation of, say, an erroneously high value for the cross section at a given energy automatically leads to the calculation of an erroneously low cross section at the next higher energy. With a sufficient number of points to determine a smooth curve, the area under this curve is relatively insensitive to the large individual errors on the points. We estimate the uncertainty in the integrated cross sections given in this paper to be about 20 percent.

Peak Cross Section and Energy at Peak

Figure 6 shows a plot of cross-section maxima as a function of mass number, the curve drawn through the measured points showing an $A^{\frac{3}{2}}$ dependence. Considering the errors quoted above on a cross-section value at a given point, this is in good agreement with the results of Montalbetti *et al.*, who find an $A^{5/3}$ relationship. No great reliability can, however, be attached to the exact value of the exponent of A. As shown in Fig. 1 of reference 12, the energy at peak cross section varies as $38.5A^{-0.186}$ in substantial agreement again with Montalbetti *et al.*, and with the predictions of Goldhaber and Teller. With a ± 1-Mev uncertainty in the position of the peaks, the statistical uncertainty in the 0.186 exponent is calculated to be ± 0.011. This result would seem to be in contradiction to the predictions of Steinwedel, Jensen, and Jensen[22] who compute an exponent of $-\frac{1}{3}$ on the basis of a hydrodynamic nuclear model.

Widths of Giant Resonances

The widths at half-maximum of the giant resonances are plotted in Fig. 7. The gradual decrease of the widths with increasing A as indicated by the straight line drawn through the experimental points of Fig. 7 is also observed by Montalbetti *et al.*, although inspection of Fig. 4 indicates some discrepancies in shapes between their results and ours. The previously reported anomalies in half-widths for the elements carbon, vanadium, columbium, and bismuth with neutron numbers of 6, 28, 52, and 126 have let us to include a study of lanthanum with 82 neutrons.

Since the sample consisted of lumps of lanthanum of irregular shape (covered with a thin oil film), absolute values of cross sections are not known, but the width of the dipole resonance is narrower than the widths for neighboring elements.

Lack of precise knowledge of the bremsstrahlung distributions and the general smoothing of data make evaluation of errors in half-widths extremely difficult. The favorable comparison of the excitation function of copper taken using a pair spectrometer to determine the bremsstrahlung spectra[23] with the methods herein employed is encouraging. Uncertainty in the spectrum should not, in fact, much influence striking differences in behavior of neighboring elements, and the ± 1-Mev errors attributed to points in Fig. 7 should be so interpreted.

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[22] Steinwedel, Jensen, and Jensen, Z. Naturforsch. **6a**, 218 (1951).

[23] V. E. Krohn and E. F. Schrader, Phys. Rev. **87**, 685 (1952).