Electro- and Photodisintegration Cross Sections of Cu^{63}

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(Received June 20, 1955)

Highly monoergic electrons from a 22-Mev betatron have been used to study the Cu^{62} activity in a pair of 2-mil copper foils separated by a 10-mil copper radiator. The ratio of the photodisintegration to electrodisintegration in the foils decreases by less than 10% in the region from 14 to 20 Mev. The measured value of F at 20 Mev is 8.6 in reasonable agreement with the extrapolation of previous measurements at higher energies and with the value of 8.38 expected on the basis of simple virtual photon calculations. Analysis of the excitation curves gave a $Cu^{63}(\gamma,n)$ cross section of 80×10^{-27} cm^2 which is 20% below most previous measurements.

INTRODUCTION

MOST investigations of (γ,n) reactions have been carried out using the bremsstrahlung spectra of electron accelerators. Activation curves are experimentally obtained by varying the kinetic energy of the electrons reaching the target (i.e., the maximum gamma-ray energy) and by observing the activity induced in samples per unit of irradiation. These activation curves are converted into cross sections with the aid of theoretical expressions for the shape of the bremsstrahlung spectrum. There is some uncertainty in the choice of a theoretical shape, associated with the questions of multiple traversals of the target by the electrons and the use of integrated (over angle) spectra as opposed to forward direction spectra.

In addition to the problem of the choice of a spectrum, the interpretation of the monitor response to arrive at unit irradiation for the various energies presents difficulties. Here one must make assumptions as to the interactions of the radiation of different energy with the material of the monitor and as to the degree of equilibrium established between the primary (gamma ray) and the secondary (electron-positron) radiation. From these assumptions one may deduce the number of photons of a particular energy band per unit of irradiation at each energy used. In the present experiment electrons free from gamma rays and of homogeneous energy are focused onto a small spot upon a copper foil stack and the emerging electrons are trapped in a Faraday cage. The monitor interpretation now presents little difficulty, since electrons of all energies give the same response to charge measuring devices, while gamma rays are not measured at all. Further, it is evident that the proper spectrum to use in interpreting this experiment is the integrated spectrum for single traversal. Multiple traversal is ruled out and the short path length from the origin of the gamma rays to their absorption acts to integrate the spectrum, at least the forward hemisphere of it. There remain, of course, experimental uncertainties, accentuated by the fact that the cross section depends on the derivatives of the activation curve and some residual doubt concerning the accuracy of the radiation expression itself.

One disadvantage of the present experiment is that to find the (γ,n) activity, the electrodisintegration activity which is due to the electrons passing through the sample must be subtracted from the total activity. Apart from energy losses and absorption, the (γ,n) activity will be proportional to the depth in the stack. Thus the intercept of the activity at zero depth will give the electrodisintegration probability while the slope of activity *versus* depth will give the product of the radiation and photodisintegration probabilities. The complication of this picture by the energy losses will be considered in more detail under the analysis of the data.

The technique of observing the variation of electron-induced radioactivity with depth in a stack of foils has been used previously in this laboratory.[1,2] Brown and Wilson[3] have made extensive measurements with stacks of foils of Cu, Zn, Ag, and Ta in order to investigate the nature of the interaction of the electromagnetic field with these nuclei. Their results are expressed in terms of the ratio of the bremsstrahlung induced activity to that produced directly by the electrons. The present work extends these results to lower energies for Cu.

Since the work was started, Berman and Brown,[4] hereafter referred to as BB, have reported on the measurements of the (γ,n) and $(\gamma,2n)$ cross sections in Cu^{63} using electron beam techniques very similar to that described here. Their work is done with less energy resolution but extends up to 36 Mev.

EXPERIMENTAL ARRANGEMENT AND PROCEDURE

Absolute and relative measurements were made of the activity induced in a stack of copper foils irradiated by monoenergetic electrons at half-Mev intervals from the Cu^{63} (γ,n) threshold to 22 Mev.

The stack consisted of an entrance and exit detector (whose activity was measured after irradiation) with or without a 10 mil radiator between the detector foils. The entrance and exit detectors were about 2 mils thick (true thickness 46.9 mg/cm^2) and 1 in.$\times\frac{3}{4}$ in. in dimension.

Irradiations employed the external electron beam of

[1] Skaggs, Laughlin, Hanson, and Orlin, Phys. Rev. **73**, 420 (1948).
[2] L. H. Lanzl and A. O. Hanson, Phys. Rev. **83**, 951 (1951).
[3] K. L. Brown and R. Wilson, Phys. Rev. **93**, 443 (1954).
[4] A. I. Berman and K. L. Brown, Phys. Rev. **96**, 83 (1954).

the 22-Mev betatron. A permendur peeler was used to increase the useful maximum energy of the electron beam. The emerging beam was collimated, and removed to a distance of about 10 feet where it was refocused by a magnetic lens to concentrate the electrons relative to gamma rays and neutrons. The focusing current was adjusted visually and photographically to bring the beam to a line focus (about $\frac{1}{8}$ in. by $\frac{3}{8}$ in. in dimensions) at the point where the copper foil stack was to be placed.

The foil stack lay deep in the mouth of a brass lined, aluminum Faraday cage (Fig. 1). The space in and around the Faraday cage and the foils was continuous with the vacuum of the betatron donut, so that prior to passing through the foils, and being trapped in the Faraday cage, the electrons were not retarded or scattered by any windows.

The charge collected by the Faraday cage passed to a vibrating reed electrometer which measured the voltage on a condenser shunted by a resistance so as to relax with the same time constant as the Cu^{62} activity. The voltage on the condenser (from a few tenths volt to one volt on one microfarad) measured on a potentiometer immediately after the betatron was turned off was thus proportional to the integrated exposure corrected for decay up to that time.

Various tests were made as to the performance of the equipment used. For example, to demonstrate that only electrons were being detected by the Faraday cage, the beam was blocked by aluminum of thickness calculated to stop the electrons. It was observed that the condenser then charged at less than one ten thousandth of its previous rate. Placing lead in the bottom of the aluminum Faraday cage to produce more bremsstrahlung from the electron beam showed no effect on the ratio of foil activity to measured irradiation.

Brass liners were added to the inside of the chamber until no effect due to backing the copper foils with material to scatter the emerging beam could be detected. Absence of this effect would indicate that no appreciable part of the beam current was being lost due to scattering out or through the Faraday cage. Tests for collection of charge in the cable connecting the Faraday cage with the electrometer circuit, made by disconnecting the cable at the Faraday cage and leaving it in place, were negative. To determine if electrons were missing the center of the foil, or if neutron or gamma ray background was causing activity in the foils, side foils were irradiated inside the Faraday cage but out of the electron beam which showed no net count. Also a foil exposed in the usual way had a $\frac{3}{8}$-in. hole punched out of its center after irradiation and the remaining area showed a negligible count over cosmic ray background.

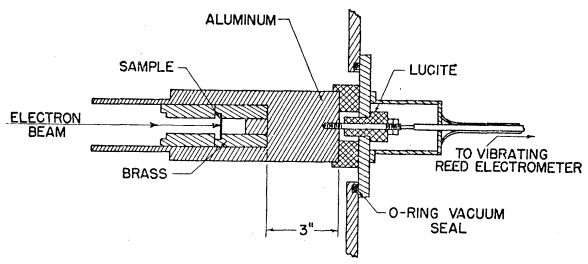

FIG. 1. Faraday cage and foil holder.

The activity in the entrance and exit detector foils were counted simultaneously between two pairs of Geiger counters during the intervals 2–6 and 7–13 minutes after the end of the irradiation. Foils were interchanged during the 6–7 minute interval so as to eliminate a small difference in counting efficiency of the pairs of counters.

The average counting efficiency of this arrangement was established at two energies by the comparison of the count of the Geiger counters on the exit foil, with the simultaneous count of the entrance foil in a 4π scintillation counter (foil sandwiched between two anthracene crystals), due account being taken of the measured ratio of exit activity to entrance activity.

The counting efficiency of the pairs of Geiger counters relative to the 4π counter was found to be nearly 55 percent.

Since the 55% measured counting efficiency was felt to be high by comparison with the 25–30% (estimated) geometrical solid angle covered by the counters, further tests were made with a $Sr\text{-}Y^{90}$ source. This source consisted of a spot ($\frac{1}{8}$ in. in diameter) of activity between two 0.002 in. copper foils of the same size and served as a counting standard for the Geiger counters. The total rate at which electrons emerged from this source was determined by means of the 4π scintillation counter as well as by a 2π gas proportional counter. The efficiency of the pairs of Geiger counters using this $Sr\text{-}Y^{90}$ source was found to agree with that using Cu^{62} sources.

It was found that the measured angular distribution was nearly of the form $\cos\theta$ where θ is measured from the normal to the plane of foil. This distribution, attributed to multiple scattering in the copper, was quantitatively sufficient to explain the observed Geiger counter efficiency.

In determining the absolute energy scale, we rely on a Cu^{63} (γ,n) threshold taken with thick gold radiators and thick copper detectors. The energy calibration in the region required for the interpretation of this experiment is based essentially upon the copper threshold which was taken to be 10.62 Mev, consistent with previous magnetic calibrations of the electron beam at 9.60 and 15.70 Mev[5] but perhaps somewhat lower than indicated by recent measurements elsewhere.[6,7] Small

[5] Goldwasser, Mills, and Hanson, Phys. Rev. **88**, 1137 (1952).
[6] M. Birnbaum, Phys. Rev. **93**, 146 (1954).
[7] Recent comparisons of the $Cu^{63}(\gamma,n)$ threshold with the $N^{14}(\gamma,n)$ and $F^{19}(\gamma,n)$ thresholds at Saskatoon indicated a value of 10.72 Mev. Katz, Penfold, and Spicer (private communication).

changes in this threshold serve only to move the energy scale of Figs. 3, 4, and 5 by a corresponding amount.

Since we intended to analyze the data using the photon difference method of analysis, we undertook to reduce the effects of energy scale drift by taking consecutively, alternating points (i.e., those 1 Mev apart) with frequent reversal of direction of traversal of the activation curve to fill in the intermediate points. Such a typical sequence of runs might be 17, 18, 19, 20, 20.5, 19.5, 18.5, 17.5, 16.5, 17 Mev for example. In all, two sets of about fifty runs each, taken three months apart, were combined to give the final data.

The observed count in each run was corrected for the observed background, dead time losses, and the decay of the Cu^{62} activity. Initially the Cu^{62} half-life was taken to be 10.0 minutes. In the subsequent analysis the half-life obtained from the observed ratio of the counts in the 2–6 minute periods to that in the 7–13 minute periods was 9.55 ± 0.15 minutes. This shorter half-life was at first attributed to a small amount of the 5 minute neutron induced Cu^{66} activity but this seemed to be excluded by the negative results with foils placed just out of the beam. This shorter half-life is in qualitative agreement with half-life of 9.73 ± 0.02 minutes reported by Berman and Brown.[4]

The absolute value of the cross section reported here is corrected to the observed half-life but nearly the same cross section is obtained by assuming a 10-minute period and enough 5-minute activity to account for the shorter period.

A further correction to the observed absolute count as measured by the scintillation counter arises from the self-absorption of the positrons in the copper sample. This absorption can be estimated as $7\frac{1}{2}\%$ by the exponential approximation used by Baker and Katz[8] to fit their measurements on Cu foils up to 20 mil thick. However, measurements carried out on $\frac{1}{2}$ mil to 2 mil foils specifically for this purpose led to a 4% absorption in the 2 mil foil which was used in this work. Subsequent calculations based on the two group diffusion theory of Bethe, Rose, and Smith using the positron energy spectrum of Cu^{62} indicated an even lower absorption of 2.5%.[9] This correction is not very sensitive to the minimum energy of positrons detected by the scintillation counter which was estimated to be 70 kev. The loss of positrons below this energy could introduce an error of less than 0.2% in the counter efficiency.

In a typical run the betatron was operated at 19.44 Mev and the charge remaining on the shunted condenser after the 10-minute irradiation was 1 microcoulomb. The two mil (46.9 mg/cm^2) entrance foil inserted between the two anthracene crystals gave a net count of 72 140 in the interval between 2 and 6 minutes after the end of the irradiation. This would be 21.1% of the total count on the basis of a 10-minute half-life. This observed activity in the entrance foil is a mixture of electron and bremsstrahlung induced activity which can be separated by the use of the activity of the exit foil. In this example the electron produced activity is 0.890 of total activity of the entrance foil. A discussion of this analysis is found in the next section.

[8] R. G. Baker and L. Katz, Nucleonics **11**, 14 (1953).
[9] M. B. Scott (unpublished report).

ANALYSIS OF DATA

Since the entrance and exit foils are behind different thicknesses of material, the ratio of the (γ,n) activity A_γ to the electrodisintegration activity A_e will be different in the two foils. The measured values of the activity of the entrance and exit foils, A_0 and A_1 respectively, can thus be used to separate A_γ and A_e. In order to do this accurately, however, it is necessary to allow for the energy loss and straggling of the electrons in the foil stack since the activities are strongly energy dependent.

For a given incident energy E_0, the electrodisintegration activity $A_e(E_0,x)$ at a depth x was calculated in terms of a linear attenuation coefficient $k(E_0)$ as defined by the relation,

$$A_e(E_0,x)=A_e(E_0,0)[1-k(E_0)x]$$
$$=\int_{E_t}^{E_0} A_e(E)P(x,E_0,E)dE, \quad (1)$$

where E_t is the (γ,n) threshold energy, E is the energy of an electron at a depth x whose incident energy was E_0. $A_e(E_0,0)$ or $A_e(E)$ is the measured electrodisintegration activity per unit thickness as obtained from the activity in the entrance foil extrapolated to zero thickness. $P(x,E_0,E)$ is the probability of finding an electron at a depth x having an energy E in a small unit energy interval. Since the energy loss distribution at a depth x can be represented by a group around the most probable energy loss having an half-width at half-maximum of about 0.1 of the energy loss, it was found unnecessary to calculate the effect of this distribution near the most probable energy in detail. On the other hand it was found that it was necessary to consider large individual energy losses arising from production of bremsstrahlung and from electron-electron collisions. The function P was therefore taken to be,

$$P(x,E_0,E)=\alpha\left(B_0+\frac{B_1}{E_p-E}+\frac{C}{(E_p-E)^2}\right). \quad (2)$$

The first two terms in the Eq. (2) represent the radiation straggling based on a trapezoidal approximation to the bremsstrahlung intensity curve. The third term is the familiar energy loss distribution associated with Rutherford scattering. $P(x,E_0,E)$ is taken as zero for energies E greater than a cutoff energy (E_p-E_c). This cutoff energy is chosen so that the

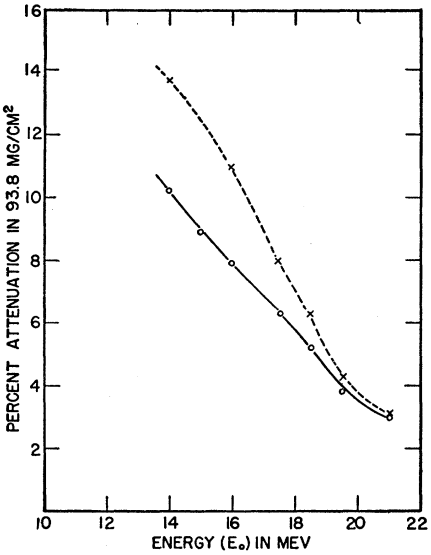

FIG. 2. Calculated attenuation coefficient $k(E_0)$ for a unit thickness of 4 mils (93.8 mg/cm²), as a function of the incident energy. The crosses represent the approximate attenuation found by assuming all electrons lost the average total energy loss.

average energy loss calculated by the relation

$$\langle E_0-E\rangle_{Av}=\frac{\int_{E_p-E_c}^{0}(E_0-E)P(x,E_0,E)dE}{\int_{E_p-E_c}^{0}P(x,E_0,E)dE} \quad (3)$$

gives the correct average total energy loss in a thickness x. α is chosen to normalize P to one electron. B_0, B_1, C and $1/\alpha$ are approximately proportional to x. The activity $A_e(E_0,0)$ was expressed in the form $a_i(E-E_i)^{ni}$ to permit analytic integration of Eq. (1) and (3). a_i, E_i, and n_i are chosen to fit segments of an experimental curve of dA_e/dE against E. Values of n_i used were 2, $1\frac{1}{2}$, 1, and $\frac{1}{2}$, and the segments were joined to make A_e and dA_e/dE continuous.

The calculated values of k for a unit thickness of 4 mils (nominal) or 93.8 mg/cm² are shown as circles in Fig. 2, together with values of k based upon the mean energy loss and the derivative dA_e/dE. It is clear from the figure that the use of the mean energy loss (neglect of straggling) is a poor approximation near the threshold.

In the linear approximation the activity per unit thickness due to electrodisintegration is

$$dA_e/dx=(1-kx)A_e(E_0,0). \quad (4)$$

Since the photodisintegration activity arises from bremsstrahlung produced at all depths from 0 to x, the effective depth is just $x/2$ and the photon induced activity can be expressed as

$$dA_\gamma/dx=[1-(kx/2)]xg(E_0,0)A_e(E_0,0), \quad (5)$$

where $g(E_0,0)$ is ratio of the photodisintegration activity behind a unit thickness to the electrodisintegration activity. In terms of these relations the activity in the entrance and exit foils are

$$A_0(E_0)=\int_0^t\left(\frac{dA_e}{dx}+\frac{dA_\gamma}{dx}\right)dx$$

$$=\left(1+\frac{g-k}{2}t-\frac{gkt^2}{6}\right)A_e(E_0)t. \quad (6)$$

$$A_1(E_0)=\int_{T+t}^{T+2t}\left(\frac{dA_e}{dx}+\frac{dA_\gamma}{dx}\right)dx$$

$$=\left\{1+\frac{g-k}{2}(3t+2T)\right.$$

$$\left.-\frac{gk}{6}(7t^2+9tT+3T^2)\right\}A_e(E_0)t, \quad (7)$$

where t is the thickness of the detector foils and T is the thickness of the radiator foil. These two equations are solved for the unknowns A_e and g. The activity due to electrodisintegration per unit thickness is

$$A_e(E_0)=\frac{A_0(E_0)}{1+\frac{(g-k)t}{2}-\frac{gkt^2}{6}}. \quad (8)$$

An idea of the importance of the various terms is gained by noting that for thickness in units of 4 mils t, T, g, and k have magnitudes of 0.5, 5, 0.5, and 0.1, respectively.

The (γ,n) activity per unit thickness squared is

$$A_\gamma(E_0)=g(E_0)A_e(E_0)=N_{Cu^{63}}\int_0^{E_0}\phi\sigma(\gamma,n)dE_\gamma, \quad (9)$$

where ϕ is the radiation cross section of Bethe-Heitler for a thin target. The cross section for electrodisintegration is simply $A_e/N_{Cu^{63}}$, while $\sigma(\gamma,n)$ can be found from the equation above by the usual photon difference method.[10] The appropriate radiation spectrum for this purpose is one integrated over angle and normalized to one electron.

In order to determine the mode by which the nucleus is excited (namely electric dipole, magnetic dipole or electric quadrupole modes),[11] Blair pointed out that it is convenient to form the ratio

$$F=\frac{A_\gamma}{A_eNr_0^2Z(Z+1)}. \quad (10)$$

[10] L. Katz and A. G. W. Cameron, Can. J. Phys. **29**, 518 (1951).
[11] J. S. Blair, Phys. Rev. **75**, 907 (1949).

This ratio as averaged over the observed $\sigma(\gamma,n)$ for Cu^{63} has been calculated and is presented in the work of Brown and Wilson.[3] Their calculation has been extended to lower energies in the present work. The value of F in terms of the quantities previously defined is simply $F=16.28g$, where g is the corrected ratio for a thickness of 93.8 mg/cm^2.

RESULTS AND DISCUSSION

In the simplest approximation of no energy loss the difference between the activity in the exit foil and the entrance foil is simply that produced by bremsstrahlung generated in the entrance and radiator foils. In order to obtain sufficient bremsstrahlung produced activity for accurate analysis it was necessary to use a radiator about 10 mils thick. In this case the corrections for energy loss are rather large and it is important that these be made accurately. The ratio of the observed activity in the exit foil to that in the entrance foil is shown in Fig. 3. The upper set of corrected points assumes all electrons in the material lose energy uniformly and that the energy at any depth in the foil is just the initial energy reduced by the average total energy loss due to ionization and radiation. The upper straight line and the open circles in the figure represent the same ratio after correcting for the energy loss in the material using the energy loss spectrum in the manner previously described. It is apparent that the points near threshold are particularly sensitive to different treatments of the energy loss. The normalized ratio of A_γ to A_e is shown as F in Fig. 4. It can be seen that this ratio at 21 Mev is not very sensitive to the manner in which the energy loss correction is made. The values observed are reasonably consistent with the

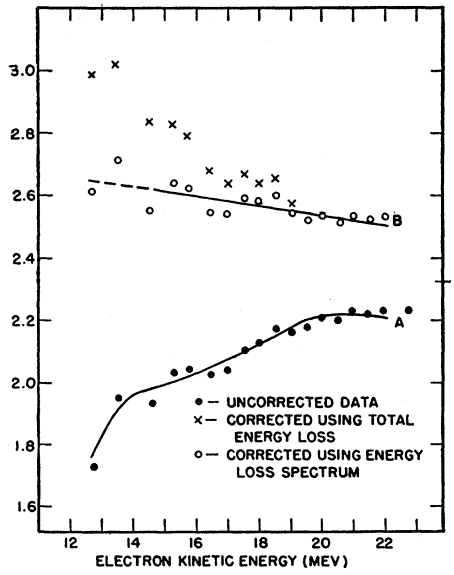

FIG. 3. Ratios of the activities in the exit foils to those in the entrance foils. The crosses represent the ratios as corrected assuming all electrons lost the average total energy loss.

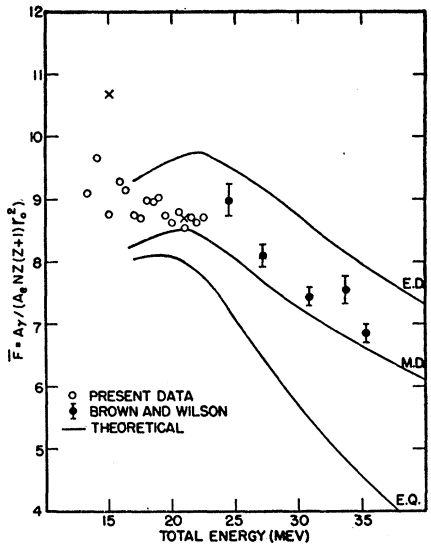

FIG. 4. Experimental values of F as obtained from the corrected values of the exit to entrance ratio. The two crosses are taken from the data which were corrected on the basis of the uniform energy loss used in Fig. 3. The solid lines represent the theoretical values of F for excitation of electric dipole, magnetic dipole, and electric quadrupole modes respectively.

extrapolation of those found by Brown and Wilson. As pointed out by them the experimental points lie closest to the theoretical values for magnetic dipole mode of absorption, but that it is more likely that these results point to a small admixture of electric quadrupole absorption with the predominant giant electric dipole resonance. Although it would be of interest to look for a change in the ratio at energies below the giant resonance, pointing more specifically to the type of interaction which is predominant in that region, it is unfortunate that the ratio is not sensitive to the mode of excitation in this region. It is also the region in which experimental measurements as well as the theoretical calculations are most unreliable.

The electrodisintegration cross section is determined with good accuracy since it depends largely on the activity in the first foil. This cross section is shown as curve 1 of Fig. 5.

The photodisintegration cross section is obtained from the corrected photon induced activity A_γ by the photon difference method. This photon-induced activity is proportional to the product of $\sigma(e,e'n)$ and F as shown in Fig. 4. Since F does not vary much in the energy region used in this work it is interesting to note that in the region around 20 Mev $\sigma(e,e'n)$ also represents the photon induced activity behind 0.175 g/cm^2 of copper. When increased to represent the corrected activity behind 0.3325 g/cm^2 as used by BB we get good agreement with the value of 3.3×10^{-28} indicated in Fig. 4 of their paper.

The photoneutron cross section is obtained from the activity A_γ by the photon difference method using the

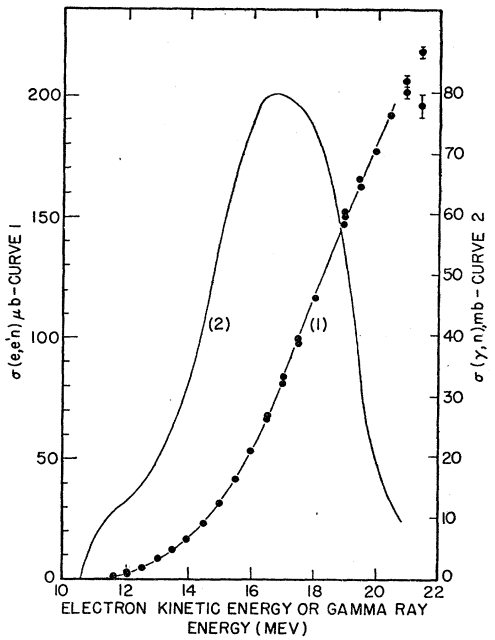

FIG. 5. The electrodisintegration activity as a function of the kinetic energy of the electron, normalized so as to represent the electrodisintegration cross section, is shown as curve 1. The statistical errors are less than the size of the points except where they are shown. The photodisintegration activity is proportional to the product of curve 1 by F as given in Fig. 4. The (γ,n) cross section as obtained from this product by the photon difference method is shown as curve 2.

total photon spectrum of Bethe and Heitler.[12] This cross section is shown as curve 2 of Fig. 5. This cross section includes the following corrections in addition to those for energy loss: (1) photon absorption in the stack (+1%), (2) multiple scattering in the stack (−2%), (3) Cu^{63} K capture transitions not detected in scintillation counter (+2%), (4) extrapolation of total count to zero thickness from 0.002 inch (+4%). The extraction of the electron beam became less reliable at higher energies so one cannot be very certain of the accuracy of the cross section at energies above 19 Mev.

It does appear to be significant that the photoneutron cross section is somewhat smaller and narrower than obtained by the earlier work using the x-ray beams. The maximum of 80 mb is less than values around 100 mb obtained in such work.[10,13–15] It is however, in better agreement with values around 85 mb obtained with 17.6-Mev rays.[16,17] The integral of the cross section at 20 Mev is 0.41 Mev barn as compared to 0.6 and greater,[16] but this work is not particularly suited for the determination of this integral because of the low maximum energy available. BB obtain a value of 0.55 Mev barn for the integrated cross section up to 34 Mev which also is below that measured previously. Their value of this integral can be obtained from the activity at the high energy without a detailed knowledge of the shape of the cross section and should therefore be more reliable than most previous values. Thus the lower maximum cross section found in this work and the lower integrated cross section found by BB agree that the previously measured cross sections are high by more than 10%.

A discrepancy of the order of 10% in the absolute cross section could arise from systematic errors in the measurement of the photon flux by means of ionization chambers.[18] It could also be that the discrepancy arises in the determination of the total number of active atoms produced. The latest measurements made use of scintillation counters which completely enclose the foils. These techniques reduce many of the systematic errors which arise in determining the effective solid angle and in correcting for absorption and scattering in the sample. In the present work the main sources of error are those associated with the absolute measurement of charge with the Faraday cage, the determination of the absolute counting rates, and the determination of F. These errors are estimated to be 3% in each case yielding an over-all estimated error of about 5%.

SUMMARY

The electrodisintegration cross section and the photodisintegration cross section have been determined in the energy range of 12 to 20 Mev. The maximum value of the (γ,n) cross section is about 20% lower than that obtained earlier with x-ray beams. The ratio of photon induced activity to electron induced activity was found to be 5 to 10% above that predicted for a magnetic dipole excitation in agreement with the trend of previous work at higher energies.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge the cooperation of T. J. Keegan in setting up and operating the betatron and the associated electron beam equipment. Han Sah was particularly helpful in the analysis of the first run and in the making of many of the measurements of the counter efficiency. We also express our thanks to Dr. J. S. Laughlin of Memorial Hospital in New York City who made a permendur peeler available to us during this period.

[12] See for example B. Rossi, *High Energy Particles* (Prentice-Hall, Inc., New York, 1952), p. 48.
[13] B. C. Diven and G. M. Almy, Phys. Rev. **80**, 407 (1950).
[14] V. E. Krohn and E. F. Schrader, Phys. Rev. **87**, 685 (1952).
[15] Karl Strauch, Ann. Rev. Nuc. Sci. **2**, 108 (1952).
[16] S. Shimizu, Mem. Coll. Sci., Univ. Kyoto **25**, 325 (1949).
[17] J. H. Carver and E. Kondaiah, Phil. Mag. **45**, 988 (1954).
[18] These questions have been discussed by L. D. Marinelli, Ann. Rev. Nuc. Sci. **3**, 249 (1953).