Cross Section for the $Ca^{40}(\gamma,3p3n)Cl^{34}$ Reaction*†

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The cross section of $Ca^{40}(\gamma,3p3n)Cl^{34}$ as a function of photon energy was determined up to 70 Mev. With a threshold for the process below 35 Mev the cross-section function was found to have a resonance-shaped peak of 0.3 millibarn at 50 Mev with a width at half-height of 6 Mev. The integrated cross section was 2.7 Mev-mb. The mode of the process is discussed on the basis of a statistical model.

INTRODUCTION

CROSS-SECTION functions of specific photonuclear reactions in the energy region just above the giant resonance absorption peak have not been studied in detail. By using radiochemical techniques the cross section for the reaction of $Ca^{40}(\gamma,3p3n)Cl^{34}$ as a function of the photon energy was determined up to 70 Mev with the Iowa State College synchrotron. This reaction was reported earlier by Schupp and Martin.[1]

Yield studies by Perlman and Friedlander[2] and others[3,4] have been made at bremmstrahlung energies of <100 Mev and some work[5–7] has been done at higher energies. These experiments indicate that the relative yields of photonuclear reactions are not strongly dependent upon the bremsstrahlung maximum energy at energies considerably above the threshold. Such results suggest a peaked cross-section function.

With the use of the Schiff[8] theoretical bremsstrahlung spectra, the flux of the x-ray beam was experimentally calibrated by three methods using (a) the reported cross-section function of the $Cu^{63}(\gamma,n)Cu^{62}$ reaction[9–12]; (b) the ionization produced in an air cavity in paraffin and its calculated response[13]; and (c) a calorimetrically calibrated ionization chamber.[14] The photonuclear yields were determined by counting the induced radioactivity which had been separated by radiochemical techniques. The cross-section functions were calculated from the activation data by the photon-difference method of Katz and Cameron.[11]

EXPERIMENTAL PROCEDURE

Redistilled calcium metal (2 g) was irradiated at various maximum bremsstrahlung energies of the synchrotron. The granular calcium metal, packed in Pyrex test tubes (10×75 mm), was centered in the x-ray beam as indicated by photographic film exposures. The calcium sample was placed behind a copper disk of equal diameter and with a thickness of about 380 mg/cm², at a distance of 17.5 cm from the bremsstrahlung target. A repeating ionization chamber, used as a monitor, was a consistency check in the activation experiments.

The energy of the electrons accelerated by the synchrotron was controlled with a recently designed integrator device.[15] In this control a pickup, annular loop was inserted above the donut chamber, and the induced voltage was integrated over time with a chopper-stabilized Miller integrator circuit. When the voltage integral attained a predetermined value, the electron orbit was expanded to strike a 5-mil tungsten target. The absolute energy calibration was believed to be well within 1.0 Mev over the energy range of these experiments on the basis of (a) the threshold checks of the $Cu^{63}(\gamma,n)$ and $Be^{9}(\gamma,p)$ reactions and (b) the maximum accelerating energy of the synchrotron. Such an energy-control device does not depend upon nuclear calibrations or upon an exact wave form for the magnetic flux.

The Cl^{34} was chemically separated by dissolving the irradiated calcium in dilute nitric acid containing chloride carrier. Under the chemical reducing conditions of the experiment, complete isotopic exchange should be assured. A silver chloride sample of about 45 mg on 6.2 cm² was precipitated from the solution and counted with an end-window Geiger counter. The reproducibility of the sample-mounting technique was experimentally shown to be about 1%. The counting efficiency of this counter-sample arrangement was determined by use of a 4π proportional counter (manufactured by Tracerlab, Incorporated). For this calibration a Cl^{34} source of high specific activity was prepared using the

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Szilard-Chalmers process by photoirradiation of carbon tetrachloride. Equal aliquots of Cl^{34} were counted (a) in the 4π counter upon a Tygon film support of less than 100 $\mu g/cm^2$ and (b) when mounted under the same conditions as in the activation experiments. The decay scheme and half-life (32.3 min) of Cl^{34} were taken from work of Nichols and Jensen[16] and the electron conversion ratio for the metastable state by Ruby and Richardson.[17]

The radioactivity induced simultaneously in the copper disk during the activation experiments was determined similarly with an end-window Geiger counter. The self-absorption and self-scattering correction was calculated by the method of Baker and Katz.[18] The counting efficiency (exclusive of self-absorption and self-scattering) for the copper was determined with the 4π counter in a manner similar to the Cl^{34}. However, in place of Cu^{62}, Y^{90}, separated from Sr^{90}, with a comparable beta-ray energy to Cu^{62} was used for the calibration. The half-life used for Cu^{62} was 9.7 min.[10]

The activation data, $Ca^{40}(\gamma,3p3n)Cl^{34}$ relative to $Cu^{63}(\gamma,n)Cu^{62}$, as a function of maximum bremsstrahlung energy are given in Fig. 1. It was assumed that the cross-section function of $Ca^{44}(\gamma,3p3n)Cl^{38}$ from the 2.1% isotope was similar to the reaction under study since small amounts of the 37-min Cl^{39} radioactivity could not be discerned. No correction was made for 2.1% electron capture[10] of the Cu^{62} but it is to be noted that other photoinduced radioactivities would largely nullify this error. The observed yield of Cu^{62} was lowered by 3% on the basis of an estimated yield for the $Cu^{65}(\gamma,3n)Cu^{62}$ reaction being 10% of the $Cu^{63}(\gamma,n)Cu^{62}$ reaction and the isotopic abundance of Cu^{65} being 29.9%.

The activation of the copper, discussed above, was also measured relative to the ionization recorded by a Victoreen 100-r chamber as a function of the maximum bremsstrahlung energy. The sensitive air volume of the

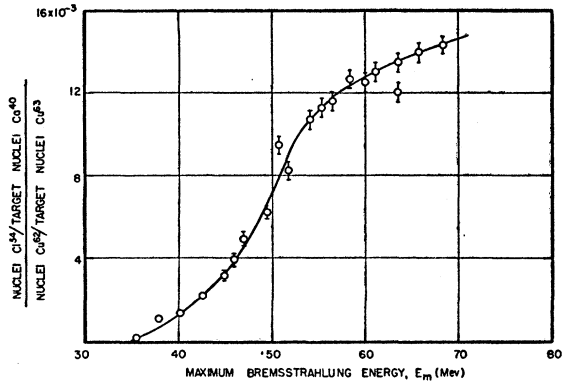

FIG. 1. Activation of $Ca^{40}(\gamma,3p3n)Cl^{34}$ relative to $Cu^{63}(\gamma,n)Cu^{62}$ as a function of bremsstrahlung energy.

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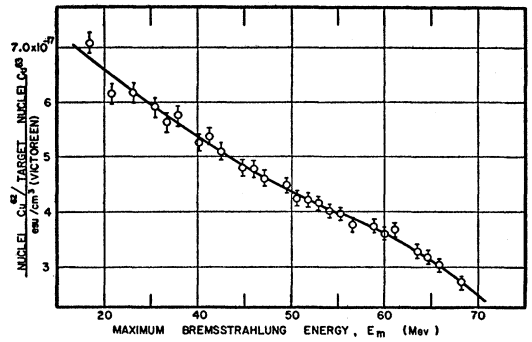

FIG. 2. Activation of $Cu^{63}(\gamma,n)Cu^{62}$ relative to ionization produced in air cavity inside paraffin.

ionization chamber was 6 inches behind the face of a 18×12×12-inch block of paraffin and at a distance of 67 cm from the x-ray source, the beam traversing the long dimension. The calcium sample was not present in these experiments. Preliminary experiments indicated that the thickness of paraffin was sufficient for equilibrium of secondary electrons with the primary photon flux. The large bulk of paraffin shielded the ionization chamber from electrons which might have been scattered from the accelerator. The activation data, $Cu^{63}(\gamma,n)Cu^{62}$ relative to esu/cm³ (Victoreen), are shown in Fig. 2.

The $Cu^{63}(\gamma,n)Cu^{62}$ reaction was also compared to the ionization measured with a Victoreen chamber in a $\frac{1}{8}$-inch thick lead cap so that the calorimetric calibration of a Victoreen thimble by Pruitt and Domen[14] could be used. A linear extrapolation was made from the data in Table I. These data, although sketchy, were used since the function appeared to be slowly changing. The center line of the uncollimated beam was obtained by scanning with the Victoreen chamber. Pruitt and Domen made their calorimetric calibration on a 25-r Victoreen chamber at a distance of about 10 m with a collimated beam.

ANALYSIS OF DATA

The cross-section functions were calculated by the photon-difference method of Katz and Cameron[11] in one-Mev intervals. The computational error from the same data was observed to be 5% when calculated by three independent computations in which different functions were smoothed. Besides the $1/r^2$ and absorption attenuations in the calcium sample, the angular

TABLE I. $Cu^{63}(\gamma,n)Cu^{62}$ relative to ionization (Victoreen chamber in lead cap at 168 cm).

Maximum bremsstrahlung energy[a] E_m (Mev)	$\left(\frac{Cu^{63}(\gamma,n)Cu^{62}/\text{nucleus } Cu^{63}}{\text{esu/cm}^3 \text{ (Victoreen)}}\right)$
45±5	$(5.01\pm0.08)\times10^{-18}$
57±5	$(4.31\pm0.14)\times10^{-18}$

[a] Accurate energy control was not possible at the time of these experiments.

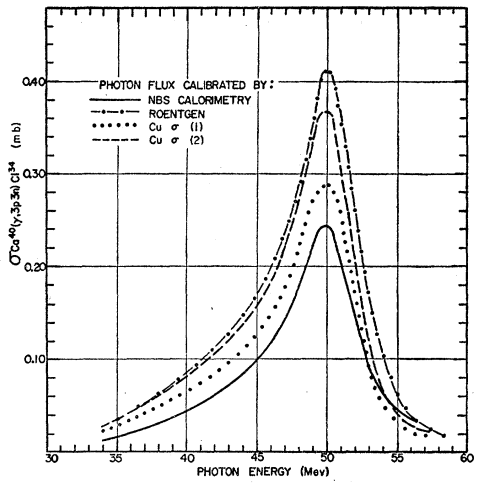

FIG. 3. Cross-section function of $Ca^{40}(\gamma,3p3n)Cl^{34}$.

distribution as a function of the electron energy was calculated using the expression by Muirhead *et al.*[19] This radial distribution has been checked experimentally with the synchrotron by Anderson and Zaffarano.[20]

If the cross-section function for the $Cu^{63}(\gamma,n)Cu^{62}$ reaction and the shape of the bremsstrahlung distribution are known, the cross-section function for the $Ca^{40}(\gamma,3p3n)Cl^{34}$ reaction may be obtained from the data of Fig. 1. The resonance cross section for copper below 23 Mev reported by Scott *et al.*[9] was used for the calculations. Their function is similar in shape and magnitude to the function obtained by Krohm and Shrader.[12] The results of the $Ca^{40}(\gamma,3p3n)Cl^{34}$ cross-section calculations based on the above copper cross-section functions are shown in Fig. 3 by the dotted curve labeled [Cuσ(1)]. For comparison, the curve [Cuσ(2)] in Fig. 3 was prepared from the $Cu^{63}(\gamma,n)Cu^{62}$ cross-section measurements of Berman and Brown[10] and the Saskatchewan group.[21]

Data in Figs. 1 and 2 give the activation function for the calcium reaction relative to the paraffin-wall chamber ionization rate. A check was possible on the data using the monitor response instead of the copper activation. The energy flux necessary to produce 1 esu/cm³ was calculated from the Gray relationship[22] by the method of Johns *et al.*[13] and Zendle.[23] The absorption coefficients were taken from the White[24] calculations. Total absorption coefficients were used for the attenuation of the beam by the doughnut, monitor, and sample. An interpolation as a function of Z was used for chemical constituents of the Pyrex glass not given in the White report. The so-called "real" or "true" absorption coefficients from the White report were used for the paraffin wall of the Victoreen ionization chamber to represent the probable energy transfer to the material by the x-ray beam. The cross-section function from these calculations is shown in Fig. 3 by the dash-dot curve.

The data from Fig. 3 and Table I, along with the experimentally determined value[14] of the energy flux necessary to produce 1 esu/cm³ (Victoreen in lead cap), were used to calculate the cross-section function. The results of the photon flux calibration by this method are shown in Fig. 3 by the solid curve.

RESULTS AND DISCUSSION

The cross-section function for $Ca^{40}(\gamma,3p3n)Cl^{34}$ was observed to be a sharply peaked function. From curve [Cuσ(1)] of Fig. 3 the maximum cross section is 0.29 millibarn at 50 Mev and the peak has a width at half-height of 6.3 Mev. The integrated cross section, $\int\sigma dE$, was 2.7 Mev-mb. This observed maximum cross section was 2% of that for the $Ca^{40}(\gamma,n)Ca^{39}$ process reported by Summers-Gill *et al.*[25] The integrated cross section was 4% of the one for the (γ,n) process.

It can be seen from Fig. 3 that all the methods of beam calibration gave similarly shaped peaks as a function of photon energy with widths of 6.0–6.5 Mev. However, the absolute magnitude of the cross sections varied significantly. It is noted that the cross-section function obtained with the paraffin-wall chamber was in fair agreement with the function dependent on a $Cu^{63}(\gamma,n)Cu^{62}$ cross section which had been obtained similarly [Cuσ(2)] in Fig. 3. The function based on the indirect calorimetry agreed fairly well with the one from the copper data of the Illinois and Case groups [Cuσ(1)]. These results imply that the $Cu^{63}(\gamma,n)Cu^{62}$ cross-section function does not have an appreciable tail above its resonance energy.

By the determination of Cl^{34} produced from the irradiated calcium target, the reaction mode(s) cannot be

TABLE II. Calculated threshold energy of $Ca^{40}(\gamma,3p3n)Cl^{34}$ reaction.

Mode	Mass threshold (Mev)	Coulombic barrier (Mev)	Threshold + barrier (Mev)
$(\gamma,\alpha d)$	26.0	17.2	42.2
$(\gamma,\alpha pn)$	28.3	17.2	45.3
$(\gamma,\alpha 2n)$[a]	31.5[b]	11.6	43.1
(γ,Li^6)	24.6	16.5	41.1

[a] $(\gamma,\alpha 2n)$ to A^{34} followed by rapid positron emission.
[b] Mass of A^{34} not experimentally determined so a calculated mass was used. [N. Metropolis and G. Reitwiesner, Atomic Energy Commission Report NP-1980, 1950 (unpublished).]

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specified, i.e., only the initial and final states are known. Listed in Table II are possible reaction modes which would produce Cl^{34} from photo-excited Ca^{40}. The mass thresholds were calculated from the mass data of Wapstra[26] and the Coulombic barrier was calculated with $r_0=1.3\times10^{-13}$ cm. As expected, detectable Cl^{34} was produced below the lowest threshold plus barrier, but well above the likely mass thresholds.

The experimental cross section for the formation of Cl^{34} was compared with the prediction of the statistical model which treats the process as particle evaporation from excited nuclei. Only ratios for cross sections can be estimated by this means without a knowledge of the probability for photon capture by the target nuclei. Therefore the ratios of the cross sections for the various modes given in Table II to the total neutron cross section, $\sigma(\gamma,n\text{-tot})$, were computed, where:

$$\sigma(\gamma,n\text{-tot})=\sigma(\gamma,n)+2\sigma(\gamma,nn)+\sigma(\gamma,np)+\sigma(\gamma,n\alpha)+\sigma(\gamma,pn)+\sigma(\gamma,\alpha n)+\cdots.$$

In this treatment the distribution, $P_i(E_i)$, for the emission of particle "i" with kinetic energy, E_i, from a nucleus with an activation energy, E, above its ground state was given by the expression,

$$P_i dE_i=\text{const}(2S_i+1)m_iE_i\sigma_i\omega_r dE_i,$$

where S_i is the spin of the particle, m_i is the mass of the particle, σ_i is the cross section for the inverse process (the capture of the particle in the indicated channel energy), and ω_r is the density of levels of the residual nucleus with an energy E_r above its ground state. The normalizing constant used in the above expression was

$$1/\text{const}=\sum_i\int_0^{E-E_b}(2S_i+1)m_iE_i\sigma_i\omega_r dE_i,$$

where E_b is the threshold energy for the emission of the particle. The summation is over all the particles which can be emitted. Values of σ_i were obtained by interpolations of the tables and graphs of Blatt and Weisskopf[27] with the parameter, $r_0=1.5\times10^{-13}$ cm. The level densities of the residual nuclei were calculated from the relation

$$(E_r)=C\exp[2(aE_r)^{\frac{1}{2}}].$$

The parameter, a, was interpolated from the table of Blatt and Weisskopf.[27] The magnitude of C is irrelevant here, and it was set proportional to 2, 1, 0.5 for odd-odd, odd-even, and even-even residual nuclei. Photon emission by a nucleus was considered negligible if a particle emission was energetically possible.

For a Ca^{40} atom with 50-Mev activation the distribution of particle emission in 2-Mev increments was first calculated. For each increment the probability distribution for the emission of a second particle was determined. These results were combined and integrated to give the residual energy distribution for the nucleus resulting from the two-particle emission. The procedure was then repeated, where necessary, to give the probability for a third particle emission below the threshold for emission of a fourth particle. The treatment indicated that $\sigma(\gamma,Li^6)$ was negligible, $\sigma(\gamma,\alpha np)/\sigma(\gamma,n\text{-tot})=0.3$, $\sigma(\gamma,\alpha nn)/\sigma(\gamma,n\text{-tot})=0.02$, and $\sigma(\gamma,\alpha d)/\sigma(\gamma,n\text{-tot})=0.06$ (for all permutations of particle emission in each case). The total neutron cross neutron for Ca^{40} has not been experimentally determined. However, the work of Jones and Terwilliger[28] on several other elements has shown this cross-section function to be slowly changing at energies above the resonance peak with a magnitude of of 10–30% of the value at the resonance peak. If 20% of the reported peak[25] is taken, a value of $Ca^{40}(\gamma,3p3n)/(\gamma,n\text{-tot})$ equal to 0.1 is indicated for the photon energy of 50 Mev. The severe limitations in accuracy for the statistical model, especially for atoms of low mass, are recognized. Furthermore, according to the shell model Ca^{40} is doubly magic. Despite these limitations, the model appears to give a fairly good prediction for the cross-section ratio. A calculation of some of the largest terms giving the ratio $\sigma(\gamma,3p3n)/\sigma(\gamma,n\text{-tot})$ at 60-Mev activation of Ca^{40} indicated a fall-off because of the competition of additional particle evaporation. However, the computed decrease was not as great as given by the cross-section curve.

On the basis of the hydrodynamic model proposed by Danos,[29] the capture cross section might be an $E1$ vibration with an internal node. However, the $Ca^{40}(\gamma,3p3n)Cl^{34}$ reaction alone exhausts the predicted capture cross section relative to the (γ,n) reaction as an $E1$ vibration in its fundamental mode.

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