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Photoneutron cross sections for bismuth-209

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Stanislaw Maria Kocimski

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I. INTRODUCTION

A study of nuclear structure is of great importance in verifying the existing theoretical models of nuclei and in providing new data for the construction of new nuclear models and nuclear interactions.

A great portion of the data comes from experimental studies of nuclear reactions. Of these, the reactions caused by γ rays are of considerable importance. These reactions are important for the following reasons. The electromagnetic interaction is reasonably well understood. Also it is relatively weak, much weaker than the nuclear interactions. This permits probing the nucleus without disturbing it too greatly.

Of great interest are the studies of simple systems like the magic nuclei. The theoretical predictions for magic nuclei are usually available, making interpretation of the results more meaningful.

Because of recent interest in the region of lead, it was decided in this work to study the cross sections for $^{209}$Bi. In addition to measuring the $(γ,n)$ cross section it was further decided to measure the cross sections for higher multiplicity reactions like $(γ,2n)$, $(γ,3n)$ etc., and compare them with the predictions of the statistical model.
In order to achieve this goal, an apparatus had to be developed capable of measuring the \((\gamma, n)\) cross section for nuclei for which the activation method as presently used in this laboratory, could not be applied.

The first step in this direction was begun by Jones (1) who built the detector with which he measured \((\gamma, n)\) and \((\gamma, 2n)\) cross sections on \(^{19}\text{F}\). However, the detector had several serious shortcomings which had to be corrected before any more precise measurements could be made.

Some of the predominant shortcomings were:

1. rather small efficiency for studying \((\gamma, 2n)\) cross sections
2. high background noise rate
3. intermediate results were not known soon enough to detect any malfunction of the system.

In the present work these shortcomings were greatly reduced. The efficiency was very high, the background noise ratio was very low, and data acquisition system gave intermediate results immediately after each run.

Subsequently the cross sections for \((\gamma, n)\), \((\gamma, 2n)\), \((\gamma, 3n)\) and \((\gamma, 4n)\) reactions were measured for \(^{209}\text{Bi}\).
II. GENERAL THEORETICAL CONSIDERATIONS

A. Introduction

The dominant feature in the interaction of $\gamma$ rays with nuclei is the giant dipole resonance. It is the broad peak which appears in the $\gamma$ ray absorption cross sections of nuclei. It appears in all nuclei; its energy and width are slowly varying functions of the mass number.

The giant dipole resonance has been the subject of intensive theoretical and experimental investigations. These investigations are not described here in detail since a number of excellent review articles have recently been published (2,3,4,5,6,7,8,9,10,11). The main interest of these investigations was to study the structure of the giant dipole resonance. Recently its splitting into components with different isospin (12) has been studied.

Most of these investigations were limited to the reactions where only one particle is emitted. Very little work had been done on reactions where more than one particle is emitted like $(\gamma,2n)$, $(\gamma,np)$, $(\gamma,3n)$ etc. The reason is that cross sections for such reactions are very small when compared with $(\gamma,n)$, and the experimental difficulties in their measurement are considerable.

The various theoretical predictions are usually based on two models, the older hydrodynamic model, and the newer shell model.
The shell model calculations are carried out using a particle-hole interaction scheme. Most of these had been made for doubly magic region like \( ^{16}O, ^{40}Ca \), because of the relative simplicity in the calculations. In the intermediate regions the calculations become too complicated to perform in detail.

Recently a great interest arose in studying the nuclei in the region of lead, \( Z=82, N=126 \). Several calculations of the cross sections for \( ^{208}Pb \) had been performed using the shell model, (13,14,15). Recent experiments were performed on \(^{208}Pb \) (16), and earlier on \(^{207}Pb, ^{208}Pb, ^{209}Bi \) (17). Theoretical models predict almost no difference in the cross sections for all nuclei in this region.

The goal in this study was to test various theoretical predictions by measuring the photonuclear cross sections for \( ^{209}Bi \).

B. Predictions for the Cross Sections

Model independent calculations lead to several so called sum rules. These rules give the values for the integrated cross sections for the photonuclear reactions.

\[
s_0 = \int \sigma(E) dE = 60 \frac{N Z}{A} \beta \text{ MeV\cdot(mb)}
\]

\( \beta \) is an adjustable parameter of the order of 1.

If we set \( \beta=1 \) then we have so called classical sum rule of Thomas Reiche and Kuhn. Other two sum rules as quoted by
Levinger (5):

\[ s_{-1} = \int \frac{\sigma(E)}{E} \, dE \]

and

\[ s_{-2} = \int \frac{\sigma(E)}{E^2} \, dE \]

for \( s_{-1} \), Levinger predicts \( s_{-1} = CA^{\frac{4}{3}} \) where \( C = 0.35 \) mb for an isotropic harmonic oscillator potential, and \( C = 0.35 \) mb for finite square well.

For \( s_{-2} \), Migdal's calculations (18) predict \( s_{-2} = 2.25A^{\frac{4}{3}} \) \( \mu b/MeV \) and Levinger (5) modifies it to \( s_{-2} = 3.5A^{\frac{4}{3}} \mu b/MeV \).

The hydrodynamical model of Goldhaber and Teller (19) explains the giant dipole resonance as due to the relative motions of two interpenetrating incompressible "fluids" of neutrons and protons. The nuclear surface remains fixed and the motion is due to the internal changes in the densities of these fluids. The restoring forces will be proportional to the gradient of the densities. The model predicts the energy of the giant resonance to be \( E_x = 80A^{-\frac{1}{3}} \) MeV. The width of the resonance is due to the number of different modes of vibrations. Since no surface vibrations are included, the only excitations are of the electric dipole nature. Shape of such a resonance can be then described by a Lorentz curve:

\[ \sigma(E) = \sigma_0 \frac{E^2 \Gamma^2}{(E^2 - E_0^2)^2 + E^2 \Gamma^2} \]
where $\Gamma$ is a width of the giant resonance.

The highly excited nucleus formed by the absorption of $\gamma$ ray may now decay in various ways. For the excitation energies below the particle threshold the decay will take place through the $\gamma$ ray emission. Above the particle threshold the nucleus may emit a neutron, proton, two neutrons, etc., depending on the excitation energy. If one considers the region above the two neutrons emission threshold, the mechanism of the reaction can be described as follows: The nucleus emits (or "boils off") the neutron losing part of its excitation energy. If the intermediate state has an excitation energy above the particle emission threshold, this state may decay again by the emission of a neutron. The statistical considerations led Blatt and Weisskopf (20) to the formula for the ratio of $(\gamma,2n)$ to $(\gamma,n)\times(\gamma,2n)$ cross sections:

$$\frac{\sigma_{2n}}{\sigma_{n} + \sigma_{2n}} = 1 - (1 + \frac{\epsilon}{\Theta}) \exp(-\frac{\epsilon}{\Theta})$$

$\epsilon$ is the excitation energy above the threshold for the $(\gamma,2n)$ process and $\Theta$ is the "nuclear temperature" governing the emission of neutrons from the intermediate nucleus.

The nuclear temperature is a slowly varying function of the excitation energy of the nucleus, which is the maximum energy of the outgoing neutrons. Blatt and Weisskopf (20) show that

$$\Theta = \left( \frac{\epsilon}{a} \right)^{\frac{1}{4}}$$
The quantity \( a \) is one of the parameters which determines the density of levels in the nucleus. Blatt and Weisskopf give that density as

\[
    w(E) = C \exp\left(2(aE)^{1/2}\right)
\]

where \( C \) is another constant ( \( C=0.01 \text{ MeV}^{-1} \) for \( A=181 \) and \( C=0.005 \text{ MeV}^{-1} \) for \( A=231 \)).

The nuclear temperature describes the energy of emitted neutrons. They will have Maxwellian distribution, and the most probable neutron energy will be \( \Theta \).

It is expected, however, that some fraction of the reactions will not go through the intermediate nucleus stage. Instead, the neutrons will be emitted in the direct process. These "direct" neutrons will not conform to Blatt and Weisskopf statistics, therefore correction should be made in the analysis of the experimental results.
III. GENERAL EXPERIMENTAL CONSIDERATIONS

A. Methods Used in the Measurements of Photonuclear Cross Sections

One common method used in the measurement of photonuclear cross sections is photo-activation.

After undergoing the photonuclear reaction where one or more neutrons or protons are emitted, the resultant nucleus is usually left in the $\beta^+$ unstable state and decays with some half-life. From the half-life time analysis a measurement can be taken to determine how many reactions took place, and from this information to obtain the cross sections. This method has been used extensively in this laboratory.

This method is limited, however, to residual nuclei which are $\beta^+$ emitters and whose life-time is between few seconds and approximately 30 minutes. Beyond these limits the experiment becomes difficult or impossible.

The closely related method is half-life analysis of metastable or excited states of residual nucleus (21).

An alternate method to the activation analysis is the direct detection method, where the protons or neutrons are detected directly from photonuclear reactions.

Since the photoneutron reactions are dominant, and its cross section measurements are the objective of this study, this paper will emphasize the method of measuring the photoneutron cross sections.
B. The Development of the Equations

Assume for the moment that we have a detector with a neutron counting efficiency $e$. We do not assume anything else about the detector. The results of the analysis should give us some requirements for this detector and for its operating conditions.

Let us first consider two extreme situations. If we pass photons one at the time through the sample, and let the detector count the outgoing neutrons, we would be able to calculate directly the ratios of differing neutron multiplicity. If on the other hand we pass a great number of photons through the sample at one burst, the detector would give us only the average number of neutrons for each beam of photons. We then could not extract ratios of different reactions taking place. In practice we have to operate at some point between these extreme positions.

Let us then assume that a burst of photons passes through the sample. Let also $R_1, R_2, R_3,$ and $R_4$ be the average number of events per beam burst of $(\gamma, n)$, $(\gamma, 2n)$, $(\gamma, 3n)$, and $(\gamma, 4n)$ nature respectively. The probability that the number of given events will take place is given by Poisson distribution:

$$P(j,k) = \frac{R_k^j e^{-R_k}}{j!}$$
\[ P(j,k) = \text{probability that } j \text{ reactions of } k \text{ type will take place (} k \text{ type means } (\gamma, kn) \). \]

So if during the period of the beam burst a number of different reactions can take place, the probability that the number of neutrons will be produced is \( r_m \), where:

\[ \begin{align*}
  r_0 &= P(0,1)P(0,2)P(0,3)P(0,4) \\
  r_1 &= P(1,1)P(0,2)P(0,3)P(0,4) \\
  r_2 &= P(2,1)P(0,2)P(0,3)P(0,4) + P(0,1)P(1,2)P(0,3)P(0,4) \\
  & \quad \text{etc.}
\end{align*} \]

in general

\[ r_m = \sum P(i,1)P(j,2)P(k,3)P(l,4) \]

where the summation extends for all indices satisfying the relation:

\[ 4i+3k+2j+i = m \]

If the detector were 100% efficient \( r_m \) would be the numbers we observe in practice. To take into account detector efficiency let us calculate the probability of observing \( n \) neutrons, \( Y_n \)

\[ \begin{align*}
  Y_0 &= r_0 + (1-\varepsilon)r_1 + (1-\varepsilon)^2r_2 + (1-\varepsilon)^3r_3 + \ldots \\
  Y_1 &= \varepsilon r_1 + 2\varepsilon(1-\varepsilon)r_2 + \ldots \\
  Y_2 &= \varepsilon^2 r_2 + \ldots \\
  & \quad \text{and in general the formula is}
\end{align*} \]

\[ Y_n = \sum_{m=n}^{\infty} \varepsilon^n(1-\varepsilon)^{m-n} \frac{m!}{(m-n)!n!} r_m \]
So at this point we have the probability of detecting \( n \) neutrons as a function of \( R_1 \) ... \( R_n \) and \( \varepsilon \).

The above derivation is a generalization of equations developed by Costa (22) and Jones (1).

The best way to invert Equation 13 is to use nonlinear least squares fitting. Before we turn to that, however, let us consider an alternate method given by Goryachev (24). In this method the moments of the probability distributions are first calculated:

\[
\overline{x} = \sum_{j=0}^{\infty} jy_j \quad 14
\]

\[
\sigma = \sum_{j=0}^{\infty} j^2y_j \quad 15
\]

From these the values of \( R_1 \) and \( R_2 \) are calculated:

\[
R_1 = (\overline{x}(\varepsilon+1) - \sigma + \overline{x}^2)/\varepsilon^2 \quad 16
\]

\[
R_2 = (\sigma - \overline{x}^2 - \overline{x})/2\varepsilon^2 \quad 17
\]

This method has the advantage of simplicity in calculations and of being in a closed form. It is felt, however, that least squares method will be more accurate, but the results of the Goryachev method for the first estimates will still be used.
C. The Least Squares Method

In order to solve the Equation 13 for $R_1$ through $R_L$, the nonlinear least squares method (NLLS) was used. The program was written in the PL/1 language and followed CURFIT routine (25) utilizing the algorithm given by Marquardt (26). This algorithm combines gradient search with the method of linearizing the fitting function by first order expansion. It permits rapid convergence from point near or far away from the minimum of $\chi^2$. To speed the convergence even more for the first estimates the results of the Goryachev method were used.

In addition to greater accuracy, the NLLS method can provide the error of the fitting. In fact, the first use of this method was to find the optimal operating conditions for the detector.

Dummy data were generated and the fitting program was used to estimate the errors in the $(\gamma, n)$ through $(\gamma, 4n)$ rates for various values of $\varepsilon$ and beam intensity (represented here as the average no. of counts/beam burst). The Figures 1 through 3 show the results of this analysis.

Several important features are immediately apparent. First, there exists an optimal beam intensity, it is about 0.3 counts/beam burst.

The second and very important feature is that in order to do meaningful analysis of reactions of higher neutron
Figure 1. Error in unfolding the $(\gamma, 2n)$ rate.
Figure 2. Error in unfolding the \((\gamma,3n)\) rate.
Figure 3. Error in unfolding the \((\gamma, 4n)\) rate.
multiplicity, the detector must have high efficiency. Any attempt to do the experiment with detector efficiency of 40% or less would result in measurements with a large possible error.

D. Corrections for the Beam Intensity Jitter

All the above analysis has been carried out assuming that beam intensity is constant, i.e., the number of photons in each beam burst is the same. This is not true, for despite all efforts beam intensity can not be held more constant than about 15% to 20%. Thus corrections for the variation must be made.

Let \( Y_n \) be some function of \( I \), the beam intensity at each pulse. Let us expand \( Y_n(I) \) in the Taylor series about \( I_0 \), some average intensity.

\[
Y_n(I) = Y_n(I_0) + (I-I_0) \frac{\partial Y_n}{\partial I} \bigg|_{I=I_0} + \frac{1}{2} (I-I_0)^2 \frac{\partial^2 Y_n}{\partial I^2} \bigg|_{I=I_0} + \cdots
\]

Let \( N(I) \) be the distribution of beam intensities for the run. The average intensity \( I_0 \) will be:

\[
I_0 = \frac{\sum N(I)}{\sum N(I)} = \frac{1}{T} \sum I \ N(I)
\]

Now the observed probabilities for detecting \( k \) neutrons will be the result of "folding" \( Y_k \) into the beam intensity distribution.

\[
\bar{Y}_k = \frac{\sum N(I) Y_k(I)}{\sum N(I)}
\]
substituting the Taylor expansion we will obtain:

\[ \bar{Y}_n = Y_n(I_0) + \frac{1}{2} \sigma^2 \frac{\partial^2 Y_n}{\partial I^2} \bigg|_{I=I_0} \]

where

\[ \sigma^2 = \frac{1}{T} \sum I^2 N(I) - I_0^2 \]

So we need to find

\[ \frac{\partial^2 Y_n}{\partial I^2} \bigg|_{I=I_0} \]

Recalling the formula for \( Y_n \) we have:

\[ \frac{\partial^2 Y_n}{\partial I^2} = \sum_{m=n}^{\infty} \epsilon^n (1-\epsilon)^{m-n} \frac{m!}{(m-n)!n!} \frac{\partial^2 r_m(I)}{\partial I^2} \]

We have then to express \( r_m \) as a function of \( I \). Recall that \( R_k \) are the average number of type \( k \) reactions per beam pulse. These numbers must be proportional to the intensity, so we can say that \( R_k = \bar{R}_k I \) where \( \bar{R}_k \) is the average number of reactions of type \( k \) per unit beam burst.

We can write now

\[ r_m(I) = \sum_{i+j+k+l} \frac{R_1 R_2 R_3 R_4}{i!j!k!l!} \epsilon^{i-j-k-l} (\bar{R}_1 + \bar{R}_2 + \bar{R}_3 + \bar{R}_4) I^{i+j+k+l} \]

where again the summation extends for all values satisfying the relation:

\[ 4l+3k+2j+i=m \]
After some algebra we arrive at the formula for

$$\frac{\partial^2 \mathbf{r}_m(I)}{\partial I^2} = \mathbf{r}_m(I) \cdot \mathbf{B}\mathbf{R}(I)$$

where $\mathbf{r}_m$ is a vector given by

$$\mathbf{r}_m = \begin{bmatrix} r_m \\ r_{m-1} \\ \vdots \\ r_{m-8} \end{bmatrix}$$

Note here that when $m$ is less or equal to eight, the components for these indices are equal to zero.

$\mathbf{B}\mathbf{R}$ is another vector given by

$$\mathbf{B}\mathbf{R} = \begin{bmatrix} (SR)^2 \\ -2\bar{R}_1(SR) \\ -2\bar{R}_2(SR)+\bar{R}_1^2 \\ -2\bar{R}_3(SR)+2\bar{R}_1\bar{R}_2 \\ -2\bar{R}_4(SR)+2\bar{R}_1\bar{R}_3+\bar{R}_2^2 \\ 2\bar{R}_1\bar{R}_4+2\bar{R}_2\bar{R}_3 \\ 2\bar{R}_2\bar{R}_4+\bar{R}_3^2 \\ 2\bar{R}_3\bar{R}_4 \\ \bar{R}_4^2 \end{bmatrix}$$

where $(SR) = \bar{R}_1+\bar{R}_2+\bar{R}_3+\bar{R}_4$

So finally, the formula for the observed probabilities of detecting $n$ neutrons in the beam burst, taking into account the beam intensity jitter, is given by

$$v_n = \sum_{m=n}^{\infty} e^{-n} (1-e)^{m-n} \frac{m!}{(m-n)!} \left( r_m + \frac{1}{2} \sigma^2 \mathbf{r}_m \cdot \mathbf{B}\mathbf{R} \right)$$
This is the formula to which we will apply non-linear least squares fitting to obtain $R_1$ through $R_4$.

E. Treatment of the Natural Background

Before the measured values of $Y_n$ will be used in the NLLS fitting, corrections for the natural background have to be made.

If the average number of counts of natural background is $B_d$, then the probability of detecting $j$ counts due to it in the counting interval, is given also by the Poisson distribution. Let $P_0$, $P_1$, $P_2$, ... $P_j$ be probabilities of detecting 0, 1, 2, ... $j$ counts from the natural background respectively.

Probability then of detecting $l$ counts in the counting interval is given by:

$$\tilde{Y}_l = \sum_{j+k=l} P_j Y_k$$

This can be written also as a matrix equation:

$$\tilde{Y} = P \cdot \tilde{Y}$$

where

$$\tilde{Y} = \begin{bmatrix} Y_0 \\ Y_1 \\ \vdots \end{bmatrix} \quad \tilde{Y} = \begin{bmatrix} \tilde{Y}_0 \\ \tilde{Y}_1 \\ \vdots \end{bmatrix} \quad P = \begin{bmatrix} P_0 & 0 & 0 & \cdots \\ P_1 & P_0 & 0 & \cdots \\ P_2 & P_1 & P_0 & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{bmatrix}$$
So \( \bar{Y} \) will be given by:

\[
\bar{Y} = e^{-1} \cdot \bar{Y}
\]

**F. Conclusions**

The above discussion suggests the following general requirements for the apparatus for the measurement of photoneutron cross sections of different multiplicities:

1. The photons should enter the sample in bursts of intensity that would produce about 0.3 neutrons per burst. The intensity should be as stable as possible.

2. The neutrons produced by the passage of the beam through the sample should be counted by the high efficiency detector (preferably close to 80%), and the recorded number stored, so that at the end of the run the distribution of different multiplicities will be available for processing.

3. There should be a monitor to record the total number of photons (total dose) which passed through the sample in the course of each run.

4. There should be another monitor registering the intensity of each beam burst as it passes the sample. The distribution of the intensities should be obtained in order to calculate the quantity used in Equation 28.
IV. DETAILS OF THE EXPERIMENTAL EQUIPMENT AND PROCEDURE

In this chapter the description will be given of all parts of the system measuring the photoneutron cross sections. The results of the discussion in the previous chapter will help to provide an understanding of the reasons for work done to achieve high performance from certain parts of the system.

A. Synchrotron

As a source of γ radiation the bremsstrahlung from the ISU Electron Synchrotron was used. The detailed description of the machine was given by Griffin (27) and Anderson (28), and for that reason will not be repeated here.

One of the main disadvantages of using this kind of source is that it does not produce monoenergetic γ rays. Instead, the bremsstrahlung has a continuous spectrum of energy up to its maximum energy called the "tip". \( E_0 \) is equal to the kinetic energy of the electrons striking the bremsstrahlung target. Experimental yield are the result of "folding" together the bremsstrahlung spectrum and the reaction cross section \( \sigma(E) \), and a special technique has to be used to "unfold" the data.
The method will be discussed further in the "Data reduction" chapter.

Another disadvantage of this particular machine is its low pulse repetition rate. The beam burst comes about 60 times a second. Recalling now that our optimal operating condition is about 0.3 counts/beam burst, an one hour run would give about 50 000 counts. This was considered to be the minimum time which should be spent to collect the data for one energy point. Since this factor could not be changed, care had to be taken to design the experiment in such a way that the possibility of making a mistake during the run was as small as possible. Such a mistake would be quite costly. It becomes obvious also, that to have intermediate results following each run would be advantageous. It would then be possible to detect any malfunctions in the system and necessary corrections could be made.

During the set-up of the experiment it was found that the synchrotron while giving a stable beam at high intensities, was rather unstable when it was tuned to the intensity which would give the desired 0.3 counts/beam burst. After eliminating many of the sources of this instability, like drifts in the R.F. system, H.V. injector system etc., a servo system was added.

Using the fact that beam intensity is a function of injection time, the servo modified the injection timing de-
pending on the difference between the current beam intensity and its preset value. This method permitted us to obtain beam stability within 15% to 20%.

B. Beam Monitoring System

Two beam monitoring systems were used in this experiment. One was used to measure the total dose of radiation in the given period of time. The other one was used to obtain the distribution of single beam intensities.

As a total dose monitor, a replica of NBS P2 ionization chamber was used. The original chamber was calibrated with precision by Pruitt and Domen (29). The total charge collected by the chamber was measured by the vibrating reed electrometer (Cary). The output voltage which was proportional to the collected charge, was read by the digital voltmeter (DVM).

As a single beam monitor a plastic scintillator mounted on a photomultiplier tube (PMT) was used. The signal from the PMT was electronically integrated and the resultant signal was read by ADC and stored by the computer. This monitor was calibrated against the ionization chamber, and was found linear within 15% throughout the working range.

Some difficulties were encountered, however, with integrating the output from the PMT. The beam which lasts for about 1 µsec has 10 nsec micro-structure, and for this reason was very difficult to integrate accurately with the
existing equipment.

C. The Neutron Detector

1. Description

The detector used in this experiment was the improved version of one used by Cook and Jones (1,23). It consisted of a cylindrical tank, about 1 m long and 1 m in diameter, made out of 1 inch thick aluminium plate. Through the center of the tank passed a stainless steel pipe 8 cm in diameter. Through this pipe the γ ray beam passed. The sample target was placed in the center of the pipe.

Twenty eight 5 inch photomultiplier tubes (EMI 9583B ) were mounted in 4 rows on the cylindrical walls of the tank. The volume of the tank was filled with an organic type liquid scintillator. The picture of the partially open detector is shown in Figure 4.

The detailed description of the tank is given in (1).
2. Principle of operation

When a high energy neutron enters the tank it scatters on hydrogen nuclei, losing energy. This process known as thermalization lasts until the neutron reaches an energy close to the energy of thermal motion of hydrogen atoms in the liquid. This process takes about 0.3 μsec. Once the neutron reaches thermal energy it can be captured by gadolinium present in the tank. The capture process is usually associated with the emission of γ rays from the excited final nucleus. These γ rays produce a light flash in the scintillator, which in turn is detected by the photomultiplier tubes.

Two important facts have to be kept in mind while designing such a detector. First, the dimension of the detector should be comparable to or larger than the average radial distance that a neutron will travel before reaching thermal energy. For this work Monte Carlo calculations (30) give this distance to be about 30 cm. Second, the material inside the tank should have a high cross section for capture of thermal neutrons. Otherwise a large portion of the neutrons will diffuse out of the tank before they will be captured.
Figure 4. The view of the partially open neutron detector.
3. Tests and procedures

In his original version Jones used a scintillator without the addition of high capture cross section elements. The neutrons were captured on hydrogen giving 2.2 MeV \( \gamma \) rays. The half-life of neutrons in the tank was 169 \( \mu \)sec and efficiency achieved was about 54\%. The natural background associated with his measurements was quite high. The foreground to background ratio was almost 1:1.

The obvious way to improve the situation was to load the scintillator with high capture cross section material. Gadolinium was chosen because of its very high thermal capture cross section (56000 barns) and its availability in this laboratory. Another advantage of using gadolinium was that the capture \( \gamma \) rays have an energy of 8.5 MeV making them easy to identify.

Some difficulties were encountered in obtaining a gadolinium compound which would readily dissolve in an organic solvent. With the help of the Metals Development Division of Ames Laboratory two compounds were found: Gd-TBP (Gd-tributylo-phosphate) and Gd-2ex-hex (Gd 2 etho hexoate). Gd-TBP was easier to dissolve and therefore was tried first. However, for some unknown reasons the entire load turned yellow after 3 months.
After recovering the gadolinium another load was made using Gd-2ex-hex.

The electronic circuits used with the tank were similar to those used by Jones. However, the data acquisition system was completely redesigned. Since PMT's had differing gains, the negative H.V. was supplied to each tube in series with a 500 K potentiometer. This was done in order to enable us to balance the gains on the phototubes. The procedure is described in detail by Jones (1).

The PMT's were arranged in two banks of 14 tubes each with adjacent tubes belonging to different banks. Electrical signals from all PMT's in the bank were added together linearly and the output signal was sent to the discriminator. Logic signals from the discriminators from each bank were then sent to the coincidence circuit. See Figure 5.

This arrangement greatly reduced the background noise. Only the signals seen by both banks simultaneously and above a certain threshold were registered. The thresholds of the discriminators and the width of the pulses (later referred to as "coincidence resolving time") were adjustable.

In order to eliminate background noise from cosmic rays the outputs from Mixers 1 and 2 were added and the output was sent to Discriminator 3. If the pulse was above a certain level (in this work, about 9 MeV), the Veto signal was gener-
ated and sent into the coincidence system. This arrangement works like a "window", that is, it passes a pulse whose height lies between lower and upper levels.

The Discriminator 1 and 2 operated in the mode where the width of the output pulse was constant, regardless of the width of input pulse. It is important to note that discriminators exist for which the discriminator will repeatedly fire for as long as there is a pulse on the input when the width of the input pulse is larger than the preset width of the output pulse. Multiple triggering, which results in false counts, can be eliminated by setting the output pulse width to the maximum width of the input pulse and then clipping the output signal to the desired width. This necessarily increases the dead time of the system and thus reduces its efficiency. This problem was encountered by Jones (1), but in this work a different type of discriminator was used which did not have this shortcoming.

The Discriminator 3 operated in the mode in which the output pulse was set to 200 nsec or the width of the input pulse, whichever was longer. This entire arrangement, which is a part of the data acquisition system, will be referred to in the future as the "pulse selection system" or PSS and represented as a single block for simplicity. In addition to the logic output an analog output was included which was the sum of all twenty eight photomultipliers. One of the first
Figure 5. Electronic arrangement of the Pulse Selection System.
Figure 6. Electronic arrangement for obtaining the spectra from different sources.
ELECTRONICS CONFIGURATION
PSS: PULSE SELECTION SYSTEM
Figure 7. Spectra obtained from the neutron detector from different sources.
NEUTRONS FROM Cf-252 GATED ON FISSIONS

CHANNEL

Am-Be

4.5γ RAY FROM 12C

CHANNEL

226Th

60Co

22Mo
tests performed on the detector was to identify the pulses caused by the neutrons. The arrangement as shown in Figure 6 was used. Thresholds on the discriminators were set to their lowest value and the coincidence resolving time was 100 nsec.

The spectra for the different sources are given in Figure 7. It can be seen immediately that the $\gamma$ rays from the Am-Be source are much higher in energy than those of Na and Co. They are 8.5 MeV capture $\gamma$ rays from the neutrons. This also shows that it should be rather easy to discriminate out the lower energy background. Separation is much better than in the previous work (1).

In order to measure the half-life of the neutrons in the tank the following procedure was used. An Am-Be source was mounted on a NaI(Tl) crystal and inserted inside the beam pipe in the center of the tank. The Am-Be source consists of finely powdered Am and Be. Am is the source of $\alpha$ particles which reacting with Be give neutrons:

$$^{4}\text{He} + ^{9}\text{Be} \rightarrow ^{12}\text{C} + ^{1}\text{n}$$

Carbon is a stable isotope, but about 60% of it is formed in the excited state, which decays giving a 4.5 MeV $\gamma$ ray. Therefore the detection of 4.5 MeV $\gamma$ ray is an indication that the neutron has been released. This can be used to measure the half life of the neutrons in the tank.

The arrangement as shown in Figure 8 was used. The 8 MeV $\gamma$ ray pulse was used to start the Time-to-Amplitude
Figure 8. The electronic arrangement to measure the half-life of the neutrons in the tank.
ELECTRONICS CONFIGURATION

TAC: TIME-TO-AMPLITUDE CONVERTER
T: DISCRIMINATOR
PSS: PULSE SELECTION SYSTEM
Figure 9. The time spectrum of the neutrons.
NEUTRON TIME SPECTRUM

NUMBER ($10^4$) vs TIME ($\mu$SEC)
converter, and the signal from the PSS stopped it. The spectrum is shown in Figure 9.

The life time was calculated to be $\tau=5.7$ µsec. This figure agrees with the Monte Carlo calculations performed for this tank (30).

For measurements of efficiency the $^{252}$Cf fission source was used. Californium emits on average 3.275 neutrons while undergoing spontaneous fission (31). Fission events were detected by using a surface barrier detector. The source was deposited on a flat surface next to the sensitive area of the detector. Both of them were placed in the evacuated container which could be easily inserted into the beam pipe of the tank (see Figure 10). The experimental circuit is shown in Figure 11.

The signal from the fission event was used to open the gate. The time length and delay of the gate could be varied. Scaler 1 counted the number of gate pulses, while Scaler 2 registered the number of neutrons while the gate was open.

The efficiency was calculated by dividing counts in Scaler 1 by counts in Scaler 2, subtracting the natural background, and dividing by the average number of neutrons per fission. Efficiency was then plotted together with BG/Signal ratio for different values of delay, gate width, coincidence resolving time, and the lower level discriminator setting. Plots are given in Figures 12 and 13.
Figure 10. The $^{252}$Cf source and the fission detector for measuring the neutron counting efficiency.
Figure 11. The electronic arrangement for measuring the neutron counting efficiency.
From these we can see that:

1. A delay of less than 2 μsec does not change efficiency significantly,
2. While the efficiency increases with the gate length, the BG/signal ratio becomes unfavorable above 35 μsec.

These trends are independent of LLD setting or coincidence resolving time.

The result (1) is very useful since in the actual run with the beam, some time must be allowed for the flash from the beam to decay.

It was decided therefore, that for the actual run the count gate would open 2 μsec after the beam passed through, and remain open for 30 μsec. The coincidence resolving time and the LLD setting were chosen after the tests with the beam, and were set to 25 nsec and 150 mV respectively. Since the beam occurs every 17 msec, it would be economical to measure the background while waiting for the next beam burst.

The timing sequence was set then as follows: (see Figure 14)

After the start of the sequence $T_0$ there is a 2 μsec wait period for the beam flash to die out. Then the first or "count gate" is generated and the Scaler 1 input is opened. After that there is about 5 msec wait and then one hundred 30 μsec gates or "background" gates are generated with
Figure 12. Efficiency and Signal/BG ratio versus the gate width.
Figure 13. Efficiency and Signal/BG ratio versus the time delay of gate opening.
The graph shows the efficiency and signal to background ratio as a function of delay before a 30 μsec gate.

- **Efficiency**:
  - Peaks at approximately 72% at 0 μsec delay.
  - Decreases to around 40% at 10 μsec delay.

- **Signal to Background Ratio**:
  - Starts at about 40 and decreases to around 20 at 10 μsec delay.
Scaler 2 open. The exact number of background gates is counted by Scaler 3.

The electronics arrangement which accomplishes that is given in Figure 15.

D. Experimental Arrangement

The geometry of the experiment is shown in Figure 16. The bremsstrahlung beam from the synchrotron was collimated by the tapered tungsten mortar precollimator. Beam was further collimated by the steel collimator on the front wall of the bunker, and then passed through the center pipe of the neutron detector, and two beam monitors.

The tank was enclosed in the concrete bunker to shield it from the natural and machine generated background. An additional lead wall was built around the tank to further reduce the background. The temperature inside the bunker was kept constant at 16°C to retard any chemical reactions which might take place in the scintillator. This also helped to reduce the photomultiplier noise. With the exception of preamplifiers, all the electronics was kept outside the bunker so not to affect the inside temperature.

During the early tests it had been found that the beam generates quite significant background (although much smaller than in previous work (1)). This background should not be confused with the natural background which is always present, but rather should be regarded as a contribution to the total
Figure 14. The timing sequence for the neutron detector.
START SIGNAL 30 $\mu$sec gate $\sim 7$ msec wait 100 x 30 $\mu$sec gates

To 2 $\mu$sec WAIT

SCALER 1 OPEN

SCALER 2 OPEN

INHIBIT SIGNAL ON
Figure 15. The electronic arrangement of the data acquisition system.
Figure 16. The geometry of the experiment.
TUNGSTEN TARGET

CERAMIC DONUT

TUNGSTEN COLLIMATOR

75 CM CONCRETE AND IRON

BORAX

PARAFFIN + BORAX

GD LOADED LIQUID SCINTILLATOR ~240 GALLONS

PARAFFIN + BORAX

10 CM LEAD

20 CM IRON

BEAM MONITORS

PLASTIC SCINTILLATOR

NBS P2 CHAMBER

PMT
yield from such events as photonuclear reactions in the air or steel pipe inside the tank, neutrons generated in the shielding walls etc. Therefore, two series of measurements should be made, one with the sample target placed inside the tank, and the other with the target removed. To obtain the effect of the target alone, these results should be subtracted.

It is very important also, to monitor for any changes in detector efficiency, NBS P2 chamber response, natural background etc. For these reasons all runs were divided into four categories called RUN, WBBG, STAN, NBBG.

**RUN**- "regular run" with beam and sample target inside the detector. The timing sequence was started by the signal from the fast beam monitor. At the end of the run, a fit was performed to obtain values $R_1$ and $R_2$, which were then plotted on the chart.

**WBBG**- same as RUN, the only difference being that the target was removed. This run measured machine generated background.

**STAN**- standard run. The Cf source was placed inside the tank, and a Sr source inside the ionization chamber. The purpose of this run was to check the efficiency of the system and the response of the ionization chamber.

**NBBG**- no beam background. No source inside the tank or ionization chamber. The purpose of this run was to measure
the natural background, the drift of the ionization chamber and its electronics.

For BUW and WBBG, the spectrum of the beam intensity was also taken.

The results of all the runs were typed on a teletype, and punched on a paper tape. The tape was then converted into IBM cards for further processing on an IBM 360/65 computer.

The results of each run included:

1. the distribution $Y_n$ for the run
2. the total dose of radiation as measured by the P2 chamber
3. the "spectrum" of the beam intensity measured by the plastic scintillator
4. the average rate of natural background

E. Data Acquisition System

While designing the experiment, it was decided to obtain maximum usage of the on-line computer and eliminate most, if not all, of the manual data taking. This eliminated most of the human errors in this experiment. Also, it made it possible for only one person to run the entire experiment.

Intermediate results were then available at the end of each run, making it possible to detect any malfunction in the system.

A program called NEUTRON was written in real-time assem-
bly language which accomplished this. The detailed description of the NEUTRON program and the SDS 910 computer is outside the scope of this thesis, however, a short description will follow.

The computer had 8192 words of core storage (24 bits per word), an eight microsecond cycle time and sixteen priority interrupts. Input/output devices were a paper tape reader, a paper tape punch, card reader, teletypewriter, a digital display and an oscilloscope display. Data acquisition devices including two digital voltmeters, three scalers, twenty four sense lines and two ADC's were connected to the computer through the remote interface unit. These devices were connected to parallel input registers in the remote interface which could be read directly into memory.

Sense lines were connected to the various points of the experimental arrangement. Through them it was possible to test experimentally various conditions (for example, whether the target is inside the tank, position of Cf and Sr sources, the scale on CARY-DVM, etc.). Four of the lines were connected to the control switches, later referred to as "light switches". The status of the sense lines, and CARY-DVM were read continuously by the computer in fixed time intervals. For the operator's convenience, the status of all sense lines was displayed in octal code on the digital display. Devices like scalers and ADC were read asynchronously, using two of
The computer's sixteen interrupts.

Interrupt I205 or "ADC interrupt" occurred at the end of the conversion process in ADC and signaled that the data were presented. The computer read the data (representing the beam intensity), calculated the average intensity for the last sixteen beam pulses, displayed it on the oscilloscope, and if the run was in progress stored the data in the memory.

Interrupt I206 or "single beam interrupt" caused the computer to read the three scalers and zero them, test the data for internal consistency, calculate the average number of counts in Scaler 1 for the last sixteen beam pulses, display the result on the oscilloscope and again if the run was in progress the data were stored in the memory. These two subroutines were executed every time the interrupt occurred, regardless of the type of the run, or if none of the runs was initiated. In order to start any of the runs, the operator had to select the proper combination of the light switches and then push the button causing I212 or "Start" interrupt. For example, for RUN the target had to be inside the tank, Cf and Sr sources out, Cary on the proper range etc. If all conditions were satisfied the run was started. The status of the run was displayed on the oscilloscope. The messages included type and current number of the run, elapsed time, spectrum of beam intensities, error messages etc.
If during the course of the run any error condition was detected by the computer (like unusually high beam, lack of beam, internal inconsistency of the read data etc.), the run was immediately halted and the proper error message was displayed. After the cause of the error was removed, the run could be restarted. The operator could stop the run, terminate it, or completely abort it by selecting the proper combination of light switches and pushing I213 or "Stop" interrupt button.

The parameters, like the duration of the run, the sampling time for reading the dose monitor etc., could be entered from the teletypewriter.

At the end of the run or at its termination, the stored data were typed and punched on a paper tape. The calculations were performed to fit the values of $R_1$ and $R_2$ and the results were typed and punched.

F. Data Collection Procedure

As was already mentioned, the runs were divided into four categories. The sequence in which they were run was as follows:

1. STAN which lasted for 10 minutes. A Cf source was placed inside the tank, and Sr source inside the ionization chamber. At the end of the run, the efficiency was calculated. Its value was used as an input parameter for the subsequent RUN.
2. RUN which lasted for 60 minutes. A Bi target of thickness of 2.55 g/cm² was placed inside the tank. At the end of the run the rates $R_1$ and $R_2$ were calculated and plotted on a chart.

3. WBBG which followed RUN lasted also 60 minutes. The target was removed and replaced by the empty target holder. Similarly, the rates $R_1$ and $R_2$ were calculated and the result plotted.

Following that, the energy was changed and the sequence started from STAN again. Every four energy steps an additional RUN was performed at 40 MeV and was followed by WBBG. This was done in order to check further the stability of the system.

The energies, the order of which was chosen at random, ranged from 8 MeV to 60 MeV in 1 MeV steps. This was done in order to diminish the effect of long time correlations on the yield curve.

The complete yield curve had in principle only one point per energy, but the points which significantly deviated from the rest of the curve were rerun. In all, four yield curves were taken.
V. DATA REDUCTION METHOD

The computations performed on the SDS 910 computer were by no means complete. The fit was performed only for $R_1$ and $R_2$, and corrections for the natural background and beam intensity jitter were not included. Also, at this point only the yield curves were available.

In this chapter the description will be given of the calculations to obtain the cross sections.

A. Preparation of Input Data

Multiplicities were computed on an IBM 360/65 computer for all four rates. The fit included the correction for the natural background according to Equation 31 and beam intensity jitter according to Equation 28.

The natural background was plotted for the entire experiment, and its average value of 0.0145 counts/gate was used in the calculations.

The corrections for the beam intensity jitter were incorporated in the nonlinear least squares fitting routine. The resulting yield curves were then plotted, the deviating points were checked for scientific grounds to be rejected and then the average curve was calculated. The experimental errors were then calculated for each yield point.
B. Unfolding the Cross Section

As was mentioned in an early chapter, the γ ray beam from a synchrotron is not monoenergetic. The experimental yields were then the result of "folding" the cross section with the bremsstrahlung spectrum. The expression for the yield per unit monitor response is given by:

\[ \alpha(E) = \frac{\int N(k,E) \sigma(k) dk}{F(E)} \]

where \( N(k,E) \) is the spectrum of the γ rays, given by Schiff (32)
\( \sigma(k) \) is the cross section for the photonuclear reaction
\( F(E) \) is the response function of the monitor.

The reduced yield is defined by:

\[ Y(E) = F(E) \alpha(E) = \int N(k,E) \sigma(k) dk \]

In order to obtain the cross section \( \sigma(k) \) we have to solve the integral Equation 33. Since the quantity \( Y \) is obtained experimentally and contains the experimental error, the simple numerical solution can be misleading due to violent propagation of errors when unfolding a matrix with the behavior of the bremsstrahlung. Numerous methods were proposed by several authors (33,34,35) to solve this problem. In this work the method of "least structure" developed in this laboratory by B.C. Cook (34) was used.
In order to describe the method, let us first approximate the integral Equation 33 by a matrix equation

\[ Y_i = \sum_{j=1}^{n} N_{ij} s_j \]  

where

\[ s_j = \frac{1}{\Delta E} \int_{E_j-\Delta E}^{E_j} \sigma(E) dE \]  

and

\[ N_{ij} = \int_{E_j-\Delta E}^{E_j} N(E,E_i) dE \]

The solution of the equation for the relative cross section is then obtained by multiplying by the inverse matrix. This is the method developed by Penfold and Leiss (33). In order to obtain the solution that does not oscillate violently, the smoothing has to be applied, preferably a nonsubjective one. The "least structure" method requires that the cross section be smoothed to a solution consistent with the accuracy of the yield curve. A solution is defined to be consistent with the yield curve accuracy in terms of the statistics \( \chi^2 \), i.e.

\[ \chi^2 = \sum_{i=1}^{n} \left( \frac{\sum_{j=1}^{n} N_{ij} s_j - Y_i}{\Delta Y_i} \right)^2 \]

where \( \Delta Y_i \) is the error in the yield at the ith energy. Solutions are accepted if \( \chi^2 \leq n \). There are, in fact, an infinite number of solutions to satisfy this condition. The "least structure" selects the "smoother" set of solutions which
satisfy the $\chi^2$ condition. It is done by minimizing the so-called "structure function" $S(s_j)$ defined as:

$$
S(s_j) = \sum_{j=1}^{n-1} P_j (s_{j+1} - 2s_j + s_{j-1})^2
$$

which is the weighted sum of the squares of the second differences. ($P_j$ is a weight which allows smoothing to be applied in a uniform manner.)

The problem of finding the smoothed solution can be written using variational calculus as:

$$
\lambda \delta S(s_j) + \delta \chi^2(s_j) = 0
$$

where $\lambda$ is a measure of smoothing applied. For fixed $\lambda$ the result of variation is:

$$
Y_i = \{ N_{ij} + \lambda (\Delta Y_i)^2 \delta_{ik} P_k \delta_{jq} N^{-1} S_j \} s_j
$$

or

$$
Y_i = M_{ij} s_j
$$

Hence

$$
S_j = M^{-1}_{ji} Y_i
$$

Further discussion of the least structure technique is found in (34).

The reduced yields were first calculated from the average yield curves, taking into account the absorption of
rays in the target, donut wall and air. Next, the background (WBBG) curve was subtracted from the RUN curve. The resultant curve which represented the effect of the target, was then processed by the "least structure" routine (CLSR).
VI. EXPERIMENTAL RESULTS AND DISCUSSION

The average reduced yield curves obtained by the method described in Chapter V were unfolded by the "least structure" procedure to obtain the cross sections. These calculations were also performed for the yield curves obtained without applying the corrections for the beam intensity jitter.

For the $(\gamma, 3n)$ and $(\gamma, 4n)$ reactions the large experimental uncertainties did not allow extraction of differential cross sections. Instead, an estimate for the integrated cross sections up to 58 MeV was obtained.

A. The Experimental Results

1. The $(\gamma, n)$ Cross Section

The $(\gamma, n)$ cross section curve, as obtained from the CLSR analysis is shown in Figure 17. The difference between this curve and the one obtained from the yield without the correction for the beam intensity jitter applied, was not detectable.

The slight negative undershoot of the cross section curve in the region above 19 MeV may be attributed to the effect of the shape of the resolution function. If one assumes that the true cross section at these energies is close to zero, then the negative portion of the resolution function contributes just the right amount of the negative cross section. (The shape of the resolution function depends on the
experimental errors of the yield curve, if the errors are large the resolution function tends to be broader and the negative undershoots are larger, if on the other hand, errors go to zero, the resolution function approaches the $\delta$-function shape).

The integrated cross section value is 2830±100 mb MeV. Harvey et al. (17) using the quasimonochromatic $\gamma$ rays obtained the $(\gamma,n)$ cross section curve as shown in Figure 18. His value of integrated cross section is 2170 mb x MeV which is about 30% lower than from this experiment. The same paper, however, gives an integrated cross section for $^{208}$Pb which is again about 30% lower than the result of Veyssiere et al. (16). The shape of the $(\gamma,n)$ cross section curve of this experiment agrees with the result of Harvey et al. rather well.

The cross section was fitted with a Lorentz curve corrected for the barrier transmission coefficient. The fit was performed for the points below 14.5 MeV, the $(\gamma,2n)$ reaction threshold. The values obtained for the Lorentz parameters were: $\sigma_0=565$ mb, $E_r=13.9$ MeV, $\Gamma=4.50$ MeV.

2. The $(\gamma,2n)$ cross section

Two cross section curves were deduced for the $(\gamma,2n)$ reaction from the reduced yield functions with and without corrections for the beam instabilities. In contrast to the $(\gamma,n)$ case, differences between these two curves can be seen. The "uncorrected" curve is shown in Figure 19 and the "cor-
rected" one in Figure 20.

The effect of the correction for the beam intensity jitter on the \((\gamma, 2n)\) cross section is quite large and also can be seen in the yield curves. However, with added uncertainty in establishing the value of \(\sigma\) of the beam intensity spread, no valid conclusion can be made as to the shape of the resultant curve. This is because the resolution function is very broad and severely distorts the shape of the cross section curve. It is also impossible to make any valid comparison with the statistical model predictions.

The integrated cross section value was 732\(\pm\)30 mb MeV. For comparison the \((\gamma, 2n)\) cross section curve as obtained by Harvey et al. is shown in Figure 21. The value of the integrated cross section in that work was 760 mb\(\times\)MeV, in good agreement with present work.

3. The \((\gamma, 3n)\) and \((\gamma, 4n)\) cross sections

The large experimental uncertainties in the yield curves made it impossible to obtain the differential cross sections for \((\gamma, 3n)\) and \((\gamma, 4n)\) reactions from CLSR routine. However, the values of the integrated cross sections up to 58 MeV were calculated.

First, Penfold-Leiss analysis was performed, which was equivalent to setting \(\lambda=0\) in the CLSR routine. The resulting values were added together to obtain the integrated cross section. Despite the large scatter of the points, the aver-
Figure 17. The (Y,n) cross section curve.
Figure 18. The $(\gamma,n)$ cross section curve as obtained by Harvey et al. (17).
\text{Bi}^{209}(\gamma, n)_{83}

PHOTON ENERGY (MeV) vs. CROSS SECTION (mb)
Figure 19. The "uncorrected" \((\gamma, 2n)\) cross section curve.
NO CORRECTION FOR BEAM INTENSITY JITTER

$^{209}\text{Bi} (\gamma, 2n)$

CROSS SECTION (mb)

PHOTON ENERGY (MeV)
Figure 20. The "corrected" (γ,2n) cross section curve.
$^{209}\text{Bi (}\gamma,2n)\$

CORRECTED FOR
BEAM INTENSITY JITTER

CROSS SECTION (mb)

PHOTON ENERGY (MeV)
Figure 21. The $(\gamma,2n)$ cross section curve as obtained by Harvey et al. (17).
Figure 22. The total absorption cross section curve as obtained in this experiment. The broken line is the Lorentz line with the parameters given in the text.
$^{209}\text{Bi} \ (\gamma, n) + (\gamma, np) + (\gamma, 2n)$
age value remained quite constant (less than 20% uncertainty in the integrated cross section for \((\gamma, 3n)\)). The method is explained in more detail in Appendix I. The error assignment was estimated from the scatter of the points about the mean obtained at higher energies.

For the \((\gamma, 3n)\) reaction, the value of the integrated cross section was \(172\pm 40\) mb\(\times\)MeV, and for \((\gamma, 4n)\) reaction \(220\pm 100\) mb\(\times\)MeV.

Wyckoff (36) measured the ratios of the yields for high multiplicity photoneutron reactions using the bremsstrahlung with the end point energy of 137 MeV. From his data he estimates relative integrated cross sections. Using the results of Harvey et al. (17) for normalization, he gave the values for the integrated cross section for \((\gamma, 3n)\) reaction as \(168\pm 25\) mb\(\times\)MeV, and for \((\gamma, 4n)\) reaction \(88\pm 22\) mb\(\times\)MeV.

Figure 22 shows the total photoneutron cross section as obtained in this experiment. The broken line shows the best fit to the Lorentz line with the parameters given above.

B. Discussion

The total integrated cross section \(s_0\) sum rule obtained in this experiment was \(3954\pm 150\) mb\(\times\)MeV. This result is significantly higher than the classical sum rule which gives \(s_0 = 3000\) mb MeV. In fact, the \((\gamma, n)\) integrated cross section alone, almost exhaust this sum rule. This suggests that there is significant contribution of Majorana exchange
force in the nuclear potential. The value obtained for parameter $\beta$ was $\beta=1.3$. Since this experiment extends only to 58 MeV, $\beta$ must be considered a lower limit to its true value. No evidence was obtained for a reduction in total cross section at higher energies. If the cross section remains constant above 30 MeV, then another 1000 mb x MeV would be obtained for $s_0$ between 60 and 140 MeV, corresponding to a $\beta$ of 1.65. Thus measurements to higher energies are clearly needed. Another important fact obtained from this experiment was that $(\gamma, 3n)$ and $(\gamma, 4n)$ reactions contribute quite significantly in the total cross section. Their total contribution up to 58 MeV was about 400 mb x MeV which was slightly above 10% of the total.

The measurement of the $s_{-1}$ sum rule gave the result $s_{-1}=263.6$ mb, which gave the value of parameter $C$ in Levinger's formula $C=0.21\pm0.02$ mb. This value agrees very well with the experimental results for $^{208}$Pb and $^{197}$Au of Veyssiere et al. (16) who obtained $C=0.20\pm0.02$ mb, and Lepretre et al. (37) for Rb, Sr, $^{89}$Y, $^{90}$Zr and $^{93}$Nb whose value was $C=0.18\pm0.02$ mb. These results, however, were about 30% lower than the theoretical predictions by Levinger who gave the values of $C=0.35$ mb for an isotropic harmonic potential, and $C=0.30$ mb for finite square well. An estimate was made of the contribution to the $s_{-1}$ sum rule assuming a constant cross section between 60 and 140 MeV. This estimate
gave 10 mb which was less than 5% of the total. The $s_{-1}$ sum rule is proportional to the mean square of electric dipole operator. Without correlations in the nuclear wave function the dipole operator is proportional to the nuclear radius. An $s_{-1}$ of 263.6 mb corresponds to an estimate of $<r^2>_{00}$ 30% smaller than the values deduced from high energy electron scattering. This fact suggests that the motion of nucleons is strongly anticorrelated inside the nucleus. (This anticorrelation may not come from the Pauli principle, however, since the single Slater determinants were used in the determination of the original value of mean square radius).

The measurement of the $s_{-2}$ sum rule gave the value $s_{-2}=16.6\pm1$ mb/MeV. The agreement with the result of Harvey et al. (17) who gave $s_{-2}=16.6\pm1.6$ mb/MeV, and theoretical prediction of Migdal is excellent.

The results discussed above, are summarized in Table 1.

C. Summary

The photoneutron cross sections for $^{209}$Bi had been measured using the direct neutron detection technique. By increasing the neutron counting efficiency of the detector up to 76%, it was possible to measure for the first time the absolute cross sections of high neutron multiplicity reactions (up to $(\gamma,4n)$) for Bismuth. By employing on-line computer for the data collection and control of the experiment it was possible to collect very large amount of data practically
without human error. The technique for correcting the results for the beam intensity instability has been developed. The experimental part of this method needs some improvement, however, especially in measuring the beam burst intensity.

The most serious limitation of the system was the low repetition rate of the synchrotron. With the rate of 60 pulses/sec it took on average 2.5 hours to obtain one point on the yield curve.

Clear evidence for including exchange forces is cited as well as evidence for nuclear correlation in the nucleus.
Table 1. Integrated cross sections and sum rules for $^{209}$Bi

<table>
<thead>
<tr>
<th>References</th>
<th>17</th>
<th>36</th>
<th>Present work</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_M$ (MeV)</td>
<td>28</td>
<td>137$^a$</td>
<td>58</td>
</tr>
<tr>
<td>$s$ ($\gamma$,n) (mbxMeV)</td>
<td>2170</td>
<td></td>
<td>2830±100</td>
</tr>
<tr>
<td>$s$ ($\gamma$,2n) (mbxMeV)</td>
<td>760</td>
<td></td>
<td>732±30</td>
</tr>
<tr>
<td>$s$ ($\gamma$,3n) (mbxMeV)</td>
<td></td>
<td>168±25</td>
<td>172±40</td>
</tr>
<tr>
<td>$s$ ($\gamma$,4n) (mbxMeV)</td>
<td></td>
<td>88±22</td>
<td>220±100</td>
</tr>
<tr>
<td>$s$ ($\gamma$,tot) (mbxMeV)</td>
<td>2930±290</td>
<td></td>
<td>3954±150$^b$</td>
</tr>
<tr>
<td>$60X_{NZ/A}$ (mbxMeV)</td>
<td>3000</td>
<td>3000</td>
<td>3000</td>
</tr>
<tr>
<td>$s_{-1}$ (mb)</td>
<td></td>
<td></td>
<td>263.6±25</td>
</tr>
<tr>
<td>$s_{-2}$ (mb/MeV)</td>
<td>16.6±1.7</td>
<td></td>
<td>16.6±1.0</td>
</tr>
</tbody>
</table>

$^a$In this case the measurement was performed for one energy only. The cross sections were deduced from the ratio of the yields assuming the absorption.

$^b$Error obtained from quadrature.
VII. LITERATURE CITED


30. G.E. Clark, Private communication


VIII. ACKNOWLEDGMENTS

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IX. APPENDIX

Let us rewrite Equation 34 in a vector form:

\[ \hat{Y} = N \cdot \hat{S} = N \cdot D \cdot \hat{S} \]

where \( \hat{S} \) is a vector of the integrated cross sections, that is \( \hat{S}_j \) is integrated cross section up to energy \( E_j \), and matrix \( D \) is defined as:

\[
D = \begin{pmatrix}
1 & 1 \\
-1 & 1 \\
0 & -1 & 1 \\
0 & 0 & -1 & 1 \\
\cdots & \cdots & \cdots & \cdots & \cdots
\end{pmatrix}
\]

Therefore

\[ \hat{S} = (N \cdot D)^{-1} \cdot \hat{Y} = D^{-1} \cdot N^{-1} \cdot \hat{Y} \]

or

\[ \hat{S} = D^{-1} \cdot \hat{y} \]

but

\[
D^{-1} = \begin{pmatrix}
1 & 1 \\
1 & 1 & 1 \\
1 & 1 & 1 & 1 \\
\cdots & \cdots & \cdots & \cdots & \cdots
\end{pmatrix}
\]

So in order to obtain the integrated cross section, all we have to do is to sum all the values of the differential cross section.
In practice, however, the measured yields have an error \( \delta Y \), therefore

\[
\hat{S} = D^{-1} \cdot N^{-1} \cdot (\hat{Y} + \delta \hat{Y})
\]

If we measure the average over large number of measurements:

\[
< \hat{S} > = < D^{-1} \cdot \hat{\sigma} > + D^{-1} \cdot N^{-1} \cdot < \delta \hat{Y} > + < D^{-1} \cdot \hat{\sigma} >
\]

we indeed will approach the true value of the integrated cross section with no bias. Let us now find the error matrix of the integrated cross section:

\[
V^S_{ij} = < (S_i - S_i) (S_j - S_j) > = \sum_{\alpha \beta} D^{-1}_{i \alpha} D^{-1}_{j \beta} N^{-1}_{\alpha} N^{-1}_{\beta} < \delta Y_\alpha \delta Y_\beta >
\]

\[
= \sum_{\alpha \beta} D^{-1}_{i \alpha} D^{-1}_{j \beta} N^{-1}_{\alpha} N^{-1}_{\beta} \epsilon_\beta^2
\]

since \( < \delta Y_\alpha \delta Y_\beta > = \epsilon_\beta^2 \delta_{\alpha \beta} \)

now the matrix

\[
V^S_{\alpha \alpha} = \sum_{\beta} N^{-1}_{\alpha \beta} N^{-1}_{\alpha \beta} \epsilon_\beta^2
\]

is an error matrix of a differential cross section \( s \). For the bremsstrahlung experiment this matrix is strongly anticorrelated, that is, the elements are oscillating very strongly between positive and negative values of about same magnitude. The error matrix for the integrated cross section
is given by:

\[ V_{ij}^S = \sum_{\alpha \alpha} D_i^{-1} D_j^{-1} v_{\alpha \alpha}^S \]

Since the multiplication by a matrix \( D \) is effectively the addition of elements of matrix \( V^S \), the elements of matrix \( V^S \) will be very small compared to the elements of \( V^S \). That means, that despite having large errors in estimating the values of a differential cross section, the error of the integrated cross section obtained by adding the values of differential cross section, will be very small. That also means that if we calculate the average values of the integrated cross section over several energy intervals, the resultant values will differ very little.

As an illustration let us assume that the bremsstrahlung has a constant value up to the end point energy. The \( N \) matrix can be written as:

\[ N_{ij} = \xi \text{ if } j \leq i \]
\[ N_{ij} = 0 \text{ if } j > i \]

Notice that \( N = D^{-1} \cdot \xi \)

The \( V^S \) matrix will be then: \( (\xi=1) \)

\[
V^S = \begin{bmatrix}
1 & 0 & 0 & 0 \\
-1 & 1 & 0 & 0 \\
0 & -1 & 1 & 0 \\
0 & 0 & -1 & 1 \\
\end{bmatrix}
\begin{bmatrix}
\epsilon_1^2 \\
\epsilon_2^2 \\
\epsilon_3^2 \\
\epsilon_4^2 \\
\end{bmatrix}
= \begin{bmatrix}
1 & -1 & 0 & 0 \\
0 & 1 & -1 & 0 \\
0 & 0 & 1 & -1 \\
0 & 0 & 0 & -1 \\
\end{bmatrix}
\]
The elements on the main diagonal are the errors in differential cross section. They are bigger than the errors in the yield. If now we obtain the $V^S$ matrix:

$$
V^S = \begin{pmatrix}
1 & 0 & 0 & 0 \\
1 & 1 & 0 & 0 \\
1 & 1 & 1 & 0 \\
1 & 1 & 1 & 1
\end{pmatrix}
\begin{pmatrix}
-\varepsilon_1^2 & 0 & 0 \\
-\varepsilon_2^2 + \varepsilon_3^2 & -\varepsilon_2^2 & 0 \\
0 & \varepsilon_3^2 + \varepsilon_4^2 & -\varepsilon_3^2 \\
0 & 0 & \varepsilon_3^2 + \varepsilon_4^2
\end{pmatrix}
\begin{pmatrix}
1 & 1 & 1 & 1 \\
0 & 1 & 1 & 1 \\
0 & 0 & 1 & 1 \\
0 & 0 & 0 & 1
\end{pmatrix}
$$

$$
= \begin{pmatrix}
\varepsilon_1^2 \\
\varepsilon_2^2 \\
\varepsilon_3^2 \\
\varepsilon_4^2
\end{pmatrix}
$$

The matrix is the same as for the yield. (It is not surprising since for this kind of bremsstrahlung the yield is just the integrated cross section). The errors are reduced and same as in the yield. For the real bremsstrahlung we can therefore expect that the errors will be of the order of the errors in the yield.