THE RICE INSTITUTE

Neutron Angular Distributions for \(^{13}(\alpha,n)^{16}\) and Excitation Functions of Alpha Particles Elastically Scattered from \(^{13}\)

by

Phillip Nolan Dean

A THESIS
SUBMITTED TO THE FACULTY
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE OF
MASTER OF ARTS

Houston, Texas
May, 1958
INTRODUCTION

Experimental investigation of nuclear energy levels can be carried out in a number of ways. Among these are the observation of angular distributions and excitation functions of elastically scattered particles and reaction products when the scattering and reactions proceed by way of excited states of a compound nucleus in the region of excitation where the states show up as well resolved resonances. This experiment involves the excited states of \( ^{8}_{\text{O}}^{17} \) in the range of excitation energies from about 8 to about 10 Mev formed by the alpha-particle bombardment of \( ^{6}_{\text{C}}^{13} \). The scattering and reaction processes studied were

\[
^{6}_{\text{C}}^{13} + ^{4}_{\text{He}} \rightarrow ^{8}_{\text{O}}^{17} \rightarrow ^{6}_{\text{C}}^{13} + ^{4}_{\text{He}}
\]

\[
^{6}_{\text{C}}^{13} + ^{4}_{\text{He}} \rightarrow ^{8}_{\text{O}}^{17} \rightarrow ^{8}_{\text{O}}^{16} + ^{1}_{\text{n}} + 2.20 \text{ Mev}
\]

Because \( ^{8}_{\text{O}}^{16} \) and \( ^{4}_{\text{He}} \) have zero spin and \( ^{6}_{\text{C}}^{13} \) and \( ^{1}_{\text{n}} \) have spin one-half, these processes are relatively simple to analyze in order to obtain angular momenta and parity values.

The \( ^{6}_{\text{C}}^{13}, n, ^{8}_{\text{O}}^{16} \) reaction was first studied in the alpha-particle energy range above 2 and up to 5.5 Mev by Bonner, Kraus, Marion and Schiffer. The experimental work described below consisted of my contribution to the measurement and analysis of neutron angular distributions at several resonances, and of a start toward the investigation of the elastic scattering. In the case of the \( ^{6}_{\text{C}}^{13}, n, ^{8}_{\text{O}}^{16} \) reaction, I aided in taking experimental angular distributions at 2.60, 2.69, 2.77 and 2.82 Mev, correcting the data for efficiency of the detector, and computing the coefficients of the Legendre
polynomial expressions for the angular distributions. This is described in Part I of the thesis. Part II of the thesis deals with the work on elastic scattering. This consisted of preliminary angular distributions and excitation curves of elastically scattered alpha-particles from thin solid foils of $^1_3 C$. The method of making the foils was an important part of this investigation.
I THE $^{13}(\alpha,n)\,^{16}$ REACTION

For this work the $^{4}\text{He}^+$ beam of the Rice Institute Van de Graaff accelerator was used. This gives a bombarding energy range of about 2-5 Mev. The target used was an evaporated carbon target, $20\mu g/cm^2$, enriched to 65% $^{13}C$. The counter used was an anthracene crystal, $1/4$ inch by $3/8$ inch mounted on a photomultiplier tube. The solid angle at the face of the crystal was $7.13 \times 10^{-3}$ stearadians. This type of counter was used because one gets a pulse height distribution, which allows one to accurately subtract background. Processing the data obtained from the crystal merits more discussion.

The crystal actually counts protons which have been scattered by the neutrons. Thus a correction has to be made for the energy dependence of the neutron-proton cross section. Another correction must be made due to a characteristic of the crystal itself. This is associated with the energy distribution of recoil protons in the crystal. In the energy range of this experiment the neutron-proton scattering is spherically symmetric in the center of mass system, i.e.

$$\frac{\sigma(\phi)}{\sigma_s} = \frac{1}{4\pi}$$

where $\sigma(\phi)$ is the differential scattering cross section in the center of mass system, and $\sigma_s$ is the integral scattering cross section. This means that $\frac{dN}{dE}$ is independent of $E$ from $E$ to $E_n$. In fact $\frac{dN}{dE} = C \frac{\sigma_s}{E_n}$ where $E_n$ is the neutron energy, $N$ is the number of protons scattered by the incident neutrons, and $C$ is a constant which includes such factors as
the number of incident neutrons, the number of protons per cubic centimeter in the crystal, and the neutron-proton scattering cross section. Graphically the distribution is

Experimentally the curve appears as

The variation from theory is due to several reasons. These are an edge effect due to the finite size of the crystal, causing a rise in the low pulse height region, imperfect light collection causing fluctuations, gamma rays causing an increase in the low pulse height region, and a broadening due to statistical fluctuations in light conversion in the photomultiplier. Due to the rise in the low pulse height region one cannot simply take the total count for computation, but must take a fraction. Then in an angular distribution one might take a safe fraction, usually one-half, of $E_n$ and count all of the protons above this energy. If the same fraction was taken at each angle, this would be a basis for a relative yield measurement. But here a difficulty arises.
In most organic crystals, particularly anthracene, the pulse height, or light yield, from each proton is not linearly dependent on the energy of the proton. This is because the ionization due to the proton moving through the crystal is so dense that the total light yield becomes proportional to the range of the proton, rather than the energy. The pulse height vs. energy curve is:

\[ E_p \]

Pulse Height relative to $^{137}$Cs internal conversion electron

Because of this non-linearity, one must be careful in taking the fraction mentioned, in order to assure taking the same energy-fraction rather than pulse height-fraction at each angle.

The mean maximum pulse height $p_m$ was determined as in the figure above, the pulse spectrum observed on a twenty channel pulse height analyzer with one-half volt windows. A pulse height ratio was determined for $E_n$ and $\frac{1}{2} E_n$ from the non-linear curve relating pulse height to energy. Then the pulse height relative to $\frac{1}{2} E_n$ is this ratio multiplied by $p_m$. This pulse height is denoted by $p_m$ on the above graph, $p_m = \frac{1}{2} \cdot p^n$. Then the yield above $p_m$ is used in the angular distribu-
tion. If this same fraction is taken for each \( E_\eta \) at the corresponding angle, then the angular distribution will be a function of the relative yield rather than the absolute yield. For the purposes of this experiment this is entirely satisfactory, since only the shape is important.

The analysis of the \((\alpha, \eta)\) reaction is relatively simple since the angular distribution is directly proportional to the Legendre polynomials, i.e.

\[
\Sigma(\theta) \sim \sum \left[ Z(l, J, \lambda, \lambda'; \frac{1}{2}, L) \right]^2 P_L(\cos \theta)
\]

where \( P_L(\cos \theta) \) are the Legendre polynomials. Thus the angular distributions can be predicted for different \( J \) values. The experimental angular distributions can be fitted with linear functions of the Legendre polynomials of the form

\[
W(\theta) = A_0 + A_1 P_1(\cos \theta) + A_2 P_2(\cos \theta) + A_3 P_3(\cos \theta) + \ldots
\]

where \( W(\theta) \) is the yield at the angle \( \theta \). Then these equations were written for five different angles, using the polynomials only up to \( P_4(\cos \theta) \), resulting in five equations with five constants \( A_L \). The equations were solved for the constants, resulting in the equation of the curve. This equation is then compared with the theoretical expressions for different \( J \) values. Examples of the type fit obtained are given in Figure 1. One is the 2.83 Mev resonance. The equation obtained was

\[
\Sigma(\theta)_{\text{exp.}} = P_0 - 0.5 P_1 - 1.0 P_2 - 0.1 P_3 + 0.5 P_4
\]

From the graph one would assign the 2.83 Mev level as a \( J = \frac{5}{2}^+ \) state with interference from a \( J = \frac{3}{2}^+ \) state. The 2.69 Mev
state was assigned as a $J = \frac{3}{2}^-$ state (see figure 1). The
2.60 and 2.77 MeV states contained too much interference to
be assigned $J$ values. An analysis of these assignments is
given by Schiffer, et al.²
II  THE $^{13}C(\alpha,\alpha)C^{13}$ REACTION

For this work the large volume scattering chamber was used in conjunction with the 6 Mev Van de Graaff accelerator. The chamber is described in a previous paper. It consists of a volume 30 inches in diameter and 13 inches deep with a differentially pumped entrance tube, a faraday cup and two scintillation detectors. The two detectors are movable, being mounted on concentric rotatable shafts. The angle of observation is accurate to $\pm 0.1^\circ$. The target volume is defined by slit systems of $\pm 0.5^\circ$ resolution.

The volume of the chamber, including detectors, is 150 liters. The detectors occupy about 20 liters. This leaves an operating gas volume of 130 liters. The $C^{13}$ comes as enriched methyl iodide. This would have to be converted to a gas, methane. Each sample of $C^{13}$, costing about $360, contains one gram of $C^{13}$. This would make 1.73 liters at STP. The chamber operates at a pressure of about 40 mm at 25°C. Under these conditions one gram of $C^{13}$ would produce 36.6 liters of methane. Thus 3.55 grams of $C^{13}$ would be required to fill the chamber at a cost of $1280. At a conservative flow rate of about 100 liters per hour, it would cost over $1000 per hour to operate the chamber with no gas recovery system. Even if 90% of the gas could be recovered it would still cost about $5000 to carry out the entire experiment. It was this high cost which led to the investigation of the possibility of using thin self-supporting foils. The procedure developed for making the foils is as follows.
The much more abundant and cheaper $^{12}$C was used first to determine the optimum conditions for producing as thin targets as possible. The methane gas given off by the methyl iodide under vacuum was admitted to a vacuum chamber at pressures up to two inches of mercury. A $^{195}$Ta filament one-half inch wide, one and one-half inches long and .008 inches thick was first used. It was heated white hot, at approximately fifty amperes at 1.8 volts for two seconds. The carbon cracks onto the hot filament and as the filament cools the carbon separates from it as a foil. The foil is then cut loose, its area measured, and it is weighed. The thinnest $^{12}$C foil obtained by this method was approximately 200 micrograms per square centimeter. The same procedure was used on $^{13}$C. The thinnest $^{13}$C foil obtained was weighed to be 151 micrograms per square centimeter. An excitation curve was taken, using this $^{13}$C foil. It is shown in figure 6. From this it was seen that the target was too thick. It was calculated from the stopping power of $^{13}$C to be 170 kev at 4.4 Mev. Therefore a new method of making the targets was devised.

The difficulty with the foregoing method is that the carbon has a minimum thickness at which it will separate from the filament. Consequently the carbon must be removed from the filament before it becomes thick enough to peel off. The only way to do this is to remove the filament by dissolving it. Then the thickness of the film can be controlled by varying the gas pressure in the chamber. The only lower limit
is the ability of the carbon foil to be self-supporting. Tantalum can no longer be used as a filament because it forms a carbide and is very difficult to dissolve without very vigorous chemical reactions, which would destroy the film. Nickel was used because it had a fairly high melting point and was easy to dissolve. The high melting point was needed since methyl iodide cracks at about 1000°C. A thin layer of carbon was cracked onto the nickel. The nickel was found to dissolve easily in a solution of $\text{HNO}_3$, $\text{H}_2\text{O}$ and $\text{HCl}$ of ratio 2:1:1. Slight heating of the solution made the process proceed more rapidly. The nickel was thin enough, .001 inches, such that a slow process would dissolve it in a reasonable time.

At first the nickel was dissolved, leaving the carbon foil in the acid. However, the foil could not be removed from the liquid without tearing it to pieces due to the surface tension. The problem then was to remove the foil from the liquid. This was achieved in the following manner. While the carbon is on the nickel, a backing is applied to it, leaving with no backing as small a surface as can be used in the experiment. In this way the foil, with the nickel dissolved, is left mounted on the backing and can be lifted perpendiculary from the solution. The backing chosen was a .001 inch thick piece of platinum. Platinum was used because it was not affected by the acid solution. The procedure for binding the carbon to the platinum is in itself unique. A
hole 5/16 inch in diameter was first cut in a one inch square piece of platinum. Several small holes approximately 1/32 inch in diameter were then punched in the platinum around the central hole. This was done to allow the bonding agent, polystyrene coil dope, to reach both sides of the platinum and form a sort of brad. This is necessary because the polystyrene does not form an acid proof bond with the platinum. The nickel is dissolved away leaving the carbon bonded to the platinum quite rigidly. After drying a coat of shellac is applied around the edge of the foil, forming a permanent bond. This procedure was carried out using C\textsuperscript{12}.

Since the foil made in the above manner could not be weighed, a well known resonance in the C\textsuperscript{12}(p,p)C\textsuperscript{12} reaction was observed with the foil as the target. This resonance was at 4.8 Mev and is reported by Reich, et al\textsuperscript{6}, to be 12 kev. Figure 2 shows the experimental level width found with this target. It is seen to be 14.5 kev. Using the approximation that if $\Gamma_{\text{exp}}$ is the experimental level width, $\Gamma$ the true level width and $T$ the target thickness, then

$$\Gamma_{\text{exp}}^2 = \Gamma^2 + T^2$$

the target thickness, $T$, was computed to be 8.1 kev. This is the level width for 4.8 Mev protons. It is of interest to calculate the thickness of the target in micrograms per square centimeter. To do this the stopping power was needed. It is given by the relation\textsuperscript{6}

$$-\frac{dE}{dx} = \frac{4\pi e^4 Z^2 N Z}{m_e v^2} \left[ \ln \frac{2 m_e v^2}{I} - \ln \left(1 - \frac{v^2}{c^2}\right) - \frac{v^2}{c^2} \right]$$
where \( Z \) is the charge of the incident particle, \( Z \) the charge of the target nucleus, \( N \) the number of atoms per cubic centimeter, \( m_e \) the mass of the electron, \( v \) the velocity of the incident particle and \( I \) the ionization potential of the target material. Using this relation the stopping power was found to be 166 Mev per centimeter. The target thickness is then \( 0.047 \cdot 10^{-3} \) centimeters. Multiplying by the density of carbon we get the target thickness to be 100 \( \mu \) grams per square centimeter. This is a substantial decrease in target thickness from that obtained in the first method. Even this reduced thickness can be reduced further by perhaps a third or even a half by using lower pressures for cracking.

Since the experiment is to be done with alphas the thickness of the above target was computed for a 4 Mev alpha. The velocity of a 4 Mev alpha is the same as that of a 1 Mev proton. Since the stopping power is directly proportional to \( Z^2 \) of the incident particle,

\[
\frac{-\left(\frac{dE}{dx}\right)_{\text{MeV p}}}{-\left(\frac{dE}{dx}\right)_{4 \text{ MeV } \alpha}} = \frac{1}{4}
\]

The stopping power of \( \text{C}^{12} \) for a 1 Mev proton was calculated to be 544 Mev per centimeter. Then the stopping power for a 4 Mev alpha is 2180 Mev per centimeter. This gives a target thickness of 110 kev. This is still rather thick for elastic scattering but as mentioned it could perhaps be reduced by one-half. It was not possible to make a \( \text{C}^{13} \) target by this
method and take an excitation curve due to a depletion of $^{13}\text{C}$ enriched methyl iodide.

Several excitation curves were taken with the thinnest $^{13}\text{C}$ foil made. It had a weighed thickness of 151 micrograms per square centimeter. The angles investigated were $39^\circ 0', 144^\circ 19', 155^\circ 51'$ and $167^\circ 12'$ in the center of mass system. The energy range investigated was 3.0 to 5.4 Mev. The angles were chosen such that the largest angle was $161^\circ 38'$ in the lab system. This was to prevent scattering from the target holder. Two other angles were chosen at $15^\circ$ intervals in the lab system.

One detector was locked at an angle of $30^\circ$ in the lab system in an attempt to use this counter as a monitor of the coulomb scattering. This would give information as to drift of the beam over the target, if the target thickness is not uniform. A graph of the excitation curve at this angle is given in figure 3. The Rutherford or coulomb cross-section is given by

$$\sigma_c = \left( \frac{2ZZe^2}{\sqrt{2} \mu v^2} \right)^2 \csc^2 \frac{\Theta}{2}$$

where $Z$ is the charge of the incident particle, $Z$ the charge of the target nucleus, $\mu$ the reduced mass, $v$ the velocity of the particle and $\Theta$ the center of mass scattering angle. To convert this to number scattered one must know the number incident. This is measured quite accurately by discharging with the beam a capacitor with accurately known
value charged to a precise voltage. It was assumed that all alphas with energy over 3 Mev are doubly ionized when they reach the Faraday cup and are counted. This is quite true since the alphas pass through two foils. The Rutherford scattering curve obtained is shown on figure 3. The displacement between the two curves can be explained by an inaccurately known target thickness. The target thickness used was obtained from the 4.4 Mev resonance of the excitation curve taken at \(167°2'\) center of mass angle. It was known from neutron work to be 25 kev wide. From the above curve it measured 170 kev. This gives a target thickness of 165 kev. From a comparison of the curves it is evident that the experimental curve has too much structure and varies too much in slope from the Rutherford scattering to be of any use for correcting the data for beam drift. If the counter had been set much farther forward it might have been of more use but the counting rate at small angles becomes unwieldy.

The excitation curves taken at the center of mass angles \(144°19'\), \(155°51'\), and \(167°12'\) are given in figures 4, 5 and 6. To correct the energy scale for target thickness the stopping power is needed in the energy range used. This was computed and is shown in figure 7. Also the target thickness was computed from this graph and was found to be 180 \(\mu\)grams per square centimeter. This agrees fairly well with the weighed thickness, within experimental error. The shape of the curves could not be corrected for target thickness because of the
large energy loss of the alpha in passing into the foil. The structure is evident in all the excitation curves but becomes less at the smaller angles. This is due to a broadening of the target at the smaller angles since the target is fixed perpendicular to the beam. A comparison of the curves with the \((\alpha, n)\) reaction shows that all of the resonances appear. With the thinner foils the structure should become much sharper, as is necessary for a phase shift analysis.
REFERENCES


4. J. B. Marion, Shell Development Company Publication.


Fig. 1. Angular distribution of neutrons from the reaction $^4\text{He} + ^{12}\text{C}(\alpha,n)^{16}\text{O}$ at alpha particle energies of 2.83, 2.69, 4.42 and 4.63 MeV.
Fig. 2. The 4.8 keV resonance of the $\text{C}^{12}(p,p)\text{C}^{12}$ reaction, used to determine target thickness. The experimental mean level width is 14.5 kev. The true level width is 12 kev, resulting in a target thickness of 2.1 kev for 4.8 keV protons.
Fig. 3. Elastically scattered alphas from $^{13}_C$ at a center of mass angle of 39°. The calculated curve is determined using a measured target thickness. To give a reference point, 500,000 alphas corresponds to a cross section of 300 mb/sterad. The separation of the curves can be explained by an error in target thickness and by addition of levels of the nucleus. It would perhaps be better to determine target thickness using the mean shape of the experimental curve as the Rutherford scattering.
Fig. 4. Center of mass cross section for the elastic scattering of alphas by $^{13}C$ at a center of mass angle of 155°51'. The target thickness is 151 $\mu$gms/cm$^2$. Note that the ordinant does not start at zero.
Fig. 5. Center of mass cross section for the elastic scattering of alphas by \( ^{13}_{2}C \) at a center of mass angle of 144°19'. The target thickness is 151 \( \mu g/cm^{2} \). Note that the ordinant does not start at zero.
Fig. 6. Center of mass cross section for the elastic scattering of alphas by $^{13}\text{C}$ at a center of mass angle of $167^\circ 12'$. The target thickness is 151 $\mu$gm/cm$^2$. This is the largest angle that could be used and the structure is most apparent here, due to an effectively thinner target seen by the detector.
Fig. 7. The stopping power of $^{13}$C for alphas vs. lab energy.