NUCLEAR REACTION MECHANISMS IN THE HEAVY ELEMENT REGION

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Paul F. Donovan
(Thesis)

June 1958

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June 1958

ABSTRACT

Angular distributions of the heavy recoils produced in the \((\alpha, 2n)\), \((\alpha, 3n)\), \((\alpha, 4n)\), \((\alpha, t)\), \((d, n)\), \((d, 2n)\), \((d, 3n)\), and \((d, p)\) reactions of \(^{209}\text{Bi}\) have been measured at various incident-particle energies. The mechanisms of the \((\alpha, 2n)\), \((\alpha, 3n)\), and \((d, 3n)\) reactions have been interpreted by comparison of these angular distributions with theoretical angular distributions resulting from Monte Carlo calculations based on a simple evaporation model. Further information concerning the reaction mechanisms of the \((\alpha, 2n)\), \((\alpha, 3n)\), and \((\alpha, 4n)\) reactions is derived from the results of experimental recoil range measurements. Probable mechanisms are suggested for several of the other reactions.
In general, background problems are often largely eliminated by heavy-recoil techniques. Also, if several particles are emitted in the course of the reaction, it is usually not possible to determine by study of the light particles which ones were associated with a particular nuclear event, unless nuclear emulsion or track-chamber techniques are used, in which case one is restricted to the study of charged-particle emission. On the other hand, in studying low-energy recoils, one is restricted to the use of very thin targets in order not to seriously change the energy and trajectory of the recoils. The effects under investigation are usually small; that is, the angle through which a heavy recoil is deflected when a light particle is emitted is usually not very great. Ranges of heavy recoils in the energy region under investigation are very short. If a differential range curve is to be measured, extremely thin absorbers must be used. Also, angular distribution results are often difficult to interpret. The relation between laboratory and center-of-mass coordinates may be double-valued, and in reactions in which several particles are emitted, complex problems in dynamics are encountered. In this work, results of Monte Carlo calculations based on a simple evaporation model are used to interpret the angular distributions obtained for some reactions in which several particles were emitted.
II. EXPERIMENTAL PROCEDURES

A. Angular Distribution Studies

1. Preparation of targets

The bismuth targets were prepared by vaporizing reagent grade bismuth metal\(^{14}\) on to 0.001 in. thick aluminum foil. The bismuth was vaporized from a heated tantalum filament in an evacuated bell jar; the thickness of the deposited bismuth was controlled by varying the time of passage of electric current through the filament. The aluminum foil was taped to the top of the bell jar in such a way that the distance from the hot bismuth to all parts of the aluminum foil was nearly constant.

The thickness of the deposited bismuth was determined in the following way. A measured area was cut from the bismuth-coated aluminum foil. The bismuth was dissolved in a few drops of concentrated nitric acid, and the solution transferred to a 5-ml volumetric flask. This procedure was repeated with a few more drops of nitric acid, and the foil and transfer pipette were rinsed with distilled water, which was then added to the volumetric flask. Then 2.0 ml of a 10% aqueous thiourea solution was added, producing the yellow bismuth-thiourea complex.\(^{15}\) The solution was diluted to volume, and the absorption in the region of minimum transmission (440 m\(\mu\)) was measured with a Beckman Model B spectrophotometer. The concentration of bismuth in the solution was read from a curve giving percent transmission as a function of bismuth concentration.\(^{16}\) The number of micrograms of bismuth per square centimeter of aluminum foil was then calculated. Targets of thicknesses varying from 0.6 \(\gamma/cm^2\) bismuth to 37 \(\gamma/cm^2\) were prepared in order to study the effect of target thickness on the angular distribution. Thickness determinations were made near the center and edge of each bismuth-coated foil in order to determine whether the bismuth was being deposited uniformly. In every case the variation in bismuth thickness from center to edge was found to be small.

2. Cyclotron bombardment procedures

All bombardments were done on the Crocker Laboratory 60-inch
cyclotron. The bismuth target is mounted in a recoil target assembly previously pictured by Vandenbosch. The target chamber is evacuated to 50 to 100 microns pressure during the bombardment, and the non-bismuth side of the target, which faces the incoming beam, is cooled by helium gas circulating between the target and the aluminum degrading foils that are used to vary the energy of the incident particle beam. In the recoil assembly a catcher foil, consisting of a circular piece of 0.001-in. thick aluminum foil 4.6 cm in diameter, is positioned behind the target at a distance determined by the angle it is desired to intercept. The position of the beam as it passes through the target is fixed by placing a graphite collimator with a 1/8-in. circular hole in front of the target assembly. After passing through the aluminum catcher foil, the beam is stopped in an electrically insulated water-cooled Faraday cup at the back of the target chamber. The beam current at the Faraday cup is measured by an electrometer. In these studies, beam currents of about 0.25 microampere were usually used. With much larger currents, there is a tendency for a film of foreign material to deposit on the target, which causes scattering of the recoils. Also, with high beam current there is the danger of vaporizing the bismuth target material from the aluminum foil.

3. Counting procedures

After bombardment, the aluminum catcher foil is removed from the target chamber and cut into 11 concentric rings. This is done by pressing the catcher foil against a steel cutter in a hydraulic press at 5000 psi. The range of angles that each ring subtends with respect to the target is calculated from the dimensions of the catcher foil and weight of the ring and the distance of the catcher foil from the target. In most experiments, the catcher was 3.0 to 6.0 cm from the target. A gross alpha-particle count is then made on each ring, and -- when necessary for identification of the products -- the alpha activities are resolved on an alpha-particle pulse-height analyzer. In deuteron bombardments, pulse analysis may be delayed 2 to 3 days until short-lived beta
and gamma activities induced in the aluminum catcher foil have substantially decayed, since the presence of large amounts of β-γ activity leads to poor resolution in the pulse-height analysis.

The following procedures are used for identifying and counting the various products produced by the reactions investigated.

a. Bi\(^{209}\)(\(α,2n\))At\(^{211}\); \(Q = -20.6\) Mev.\(^{18}\) At helium ion energies below about 39 Mev, pulse-height analysis shows that substantially all the alpha activity produced is due to At\(^{211}\) (~7.5 hr, \(α\))\(^{19,20,21,8}\) and Po\(^{211}\) (0.52 sec, \(α\)).\(^{22}\) The Po\(^{211}\) is formed by the electron-capture decay of At\(^{211}\) (59.1% E.C.).\(^{19}\) Since the Po\(^{211}\) is in equilibrium with the At\(^{211}\), at these energies the production of \((α,2n)\) recoils as a function of angle may be studied by simply gross alpha-counting the rings.

b. Bi\(^{209}\)(\(α,3n\))At\(^{210}\); \(Q = -27.9\) Mev. Since At\(^{210}\) (8.3 hr, E.C.)\(^{21,23}\) decays mostly by electron capture, this reaction is studied by allowing the rings to decay for several days after counting the \((α,2n)\) and \((α,4n)\) recoils. This allows the At\(^{211}\) and At\(^{209}\) to decay away, and the Po\(^{210}\) (138.3 d, \(α\))\(^{24}\) daughter of the At\(^{211}\) is gross alpha counted. Some Po\(^{210}\) is undoubtedly formed directly by the Bi\(^{209}\)(\(α,t\))Po\(^{210}\) reaction, and by decay of Bi\(^{210}\) formed by the Bi\(^{209}\)(\(α,He^3\))Bi\(^{210}\) reaction, but cross-section measurements of these reactions in the heavy-element region\(^{25,26}\) indicate that the amount of Po\(^{210}\) formed in this way should be small compared with that formed by decay of At\(^{210}\) at the energies studied.\(^8\)

c. Bi\(^{209}\)(\(α,4n\))At\(^{209}\); \(Q = -35.6\) Mev. The fraction of At\(^{209}\) (5.5 hr, 5%\(α\))\(^{27}\) present in the rings is determined by alpha-particle pulse-height analysis (Fig. 1) of each ring. The rings are also gross alpha counted to determine the absolute counting rate.

d. Bi\(^{209}\)(\(α,t\))Po\(^{210}\); \(Q = -14.8\) Mev. This reaction was studied at 28.0 Mev. At this energy no At\(^{210}\) is formed. Therefore the only other source of Po\(^{210}\) would be expected to be that formed from the decay of Bi\(^{210}\) produced by the Bi\(^{209}\)(\(α,He^3\))Bi\(^{210}\) reaction. Since the cross section for \((α,He^3)\) reactions at these energies is found to be much less than that for the \((α,t)\) reaction,\(^{25,26}\) all the Po\(^{210}\) was assumed to be
Fig. 1. Pulse-height analysis of ring 2, \( \theta \approx 8^\circ \), (solid line) and energy versus channel number (dashed line) for the reaction products of the system Bi\(^{209}\) + He\(^4\) (\(E = 46.5\) Mev).
formed directly by the (α, t) reaction. The Po\textsuperscript{210} was measured by gross alpha counting after the short-lived alpha emitters had decayed away. Owing to the low cross-section of the (α, t) reaction at this energy, a long bombardment (10 hrs) at a high beam current (1 microampere) was required to produce enough Po\textsuperscript{210} to be counted easily. This high total beam led to the production of some Po\textsuperscript{210} from trace impurities (probably lead) in the innermost rings of the aluminum foil used as the catcher. Since this activity would be produced throughout the thickness of the aluminum foil, and the Po\textsuperscript{210} coming from the (α, t) reaction is only on the front surface, the Po\textsuperscript{210} coming from (α, t) was determined by gross alpha counting both sides of the rings and taking the difference. (Po\textsuperscript{210} alpha particles do not penetrate through the thickness of the rings.)

e. Bi\textsuperscript{209}(d,n)Po\textsuperscript{210}; Q = 2.8 MeV  
Bi\textsuperscript{209}(d,p)Bi\textsuperscript{210}; Q = 2.4 MeV.

At deuteron energies below 13 Mev, substantially all the alpha activity is due to Po\textsuperscript{210} produced by the (d,n) reaction and to Po\textsuperscript{210} coming from decay of the short-lived isomer of Bi\textsuperscript{210} \((5.02 \text{ d, } 99.4\% \beta^-).\) 28,29

The rings are gross alpha counted shortly after the bombardment and again after most of the Bi\textsuperscript{210} has decayed. From these data the amounts of Bi\textsuperscript{210} and Po\textsuperscript{210} produced by the bombardment are calculated. Because of the low counting rates involved, it proved impractical to apply this procedure at higher energies, at which Po\textsuperscript{208} is formed by the (d, 3n) reaction and pulse analysis is required to observe the Po\textsuperscript{210}, which usually has a low counting rate compared with that of the Po\textsuperscript{208}.

f. Bi\textsuperscript{209}(d, 2n)Po\textsuperscript{209}; Q = -4.9 MeV.  
Bi\textsuperscript{209}(d, 3n)Po\textsuperscript{208}; Q = -11.5 Mev. Because of the long half life of Po\textsuperscript{209} \((100 \text{ yr, } \alpha)^8\) it was possible to study this reaction only at one energy, near the peak of the excitation function. Po\textsuperscript{208} \((2.93 \text{ yr, } \alpha)^30,31\) recoils were observed at deuteron energies ranging from 14.5 Mev to 23.5 Mev. These products were identified by pulse analysis of the rings (Fig. 2). Absolute counting rates were determined by gross alpha counting.
Fig. 2. Pulse-height analysis of ring 5, \( \theta \approx 19^\circ \), (solid line) and energy versus channel number (dashed line) for the reaction products of the system Bi\(^{209}\) + H\(^2\) (\(E = 15.1\) Mev).
4. **Treatment of data**

For each ring, the relative solid angle subtended was calculated from the ring dimensions. Counting rates were tabulated on a relative basis as counts per minute per unit solid angle. Plots of this quantity versus the laboratory angle in degrees were constructed. Where necessary, corrections for growth and decay during the counting period and for decay during bombardment were made.

**B. Range Studies**

1. **Preparation of targets and absorber films**

The recoil range-measurement technique described below was developed by Harvey and Wade. Bismuth targets were prepared in the manner described above for the angular distribution studies. Targets of about one microgram of bismuth per square centimeter were usually used in the recoil range studies. The absorber films used in making the differential range measurements were made of VYNS plastic. The films were prepared by stretching on a water surface in the manner described by Pate and Yaffe. The films, which were 1 inch in diameter, were mounted on aluminum frames which were spaced in slots 0.030 inch apart. The film thicknesses were usually in the range of 5 to 15 micrograms per square centimeter. Depending on the film thicknesses, a stack of 15 to 20 films was used to stop the recoils. Film thicknesses were measured by the optical-absorption method described by Pate and Yaffe, by use of a Beckman Model DU spectrophotometer at a wave length of 600 mμ. The transmission-thickness calibration curve given by Pate and Yaffe was used. Results of successive measurements made on the same film are quite reproducible, and measurements made at different areas of the same film are usually in good agreement. Three of the films were weighed on a microbalance, and the weights given by this method were in good agreement with those calculated from the transmission measurements. The film thicknesses as measured before the bombardment are probably accurate to within about 0.5 μg/cm².
III. MONTE CARLO CALCULATIONS

The application of the Monte Carlo method to nuclear reaction problems was suggested by Ulam and Von Neumann. The first calculations of this type were done by Goldberger, and were based on a nuclear cascade model. Calculations based on similar cascade models have since been carried out by several authors. The results have been in qualitative agreement with experimental data on yields and angular distributions for fast cascade particles. Meadows, Jackson, and Rudstam have used calculations of the Monte Carlo type based on an evaporation model to explain experimental cross-section data.

The Monte Carlo calculations reported in this work are the result of an effort to provide predictions based on a simple model for comparison with experimental angular distribution data on heavy recoil nuclei from reactions in which several light particles have been emitted. A simple evaporation model, based on the compound nucleus concept, has been taken as the starting point for the calculations.

In order to facilitate the calculations, several simplifying assumptions and approximations have been made. These are listed and discussed below.

1. Neutrons are the only particles emitted from the compound nucleus. Competitive de-excitation by other modes, such as proton and gamma-ray emission, are neglected as long as the nucleus is excited above the binding energy of the last neutron.

2. The free evaporation energy spectrum of the neutrons emitted from the excited nucleus, unmodified by thermodynamic requirements, is given by the expression \( P(E) \propto E e^{-E/T} \), where \( P(E) \) is the probability of evaporating a neutron of energy \( E \), \( E \) is the energy of the evaporated neutron, and \( T \) is a parameter, usually referred to as the "nuclear temperature," used to adjust the shape of the evaporation energy spectrum. The energy spectrum of the emitted neutrons leading to a particular reaction at the excitation energies investigated is so drastically changed from the form given above by the imposition of thermodynamic requirements on the outgoing neutrons, and by competition from other neutron-out reactions, that the model is not very sensitive to the exact form of the unmodified energy spectrum.
3. The angular distribution of the outgoing neutrons is isotropic in
the system of the recoiling nucleus. This provides a simple model with
which experimental angular distributions may be compared.

4. The parameter $T$ is constant throughout the evaporation chain.$^{53}$
The model does not seem to be sensitive to this assumption. Monte Carlo
calculations have been carried out in which $T$ was varied by several
tenths Mev without any appreciable effect on the angular distributions.

The following approximations have been made in order to simplify
the equation that predicts the angle of the recoil nucleus in the labor-
atory system.

5. The distance traveled by the recoiling nucleus in the laboratory
system during the evaporation process is negligible compared with the
distance from the target to the catcher foil. This approximation in-
troduces no measurable error in the calculation if the total time for
neutron evaporation is less than about $10^{-9}$ second.

6. The mass of the nucleus does not change during the evaporation
process. For Bi$^{209}$, this introduces an error of less than 1% in the
angle of the recoil if not more than four neutrons are evaporated.

7. The changes in momenta of the recoiling nucleus due to loss of
mass during the evaporation process is negligible. This allows the
momentum of the recoil nucleus in the system of the struck nucleus to
be calculated without taking into account the alteration of the system
of the recoiling nucleus by the loss of neutrons. The error in the
angle of the recoil nucleus due to this approximation is less than 2%
in the least favorable case, where the errors add for the several
neutrons.

Within the limits of the above approximations, the relation
giving the angle of the recoil nucleus in the laboratory system for
the case of three emitted neutrons is

$$\tan \phi = \frac{\sqrt{1 - \cos^2 \theta_3}}{\cos \theta_3 + \frac{P_s}{P_n}}$$

where $\phi$ is the angle of the recoil nucleus in the laboratory system.
with respect to the incident particle beam; \( p_a \) is the momentum of the incident particle; and \( p_n \) is given by the expression

\[
p_n = \left[ 2mE_1 + 2mE_2 + 2mE_3 + 4\sqrt{mE_1 \sqrt{mE_2 \cos \theta_1}} + 2 \sqrt{2mE_3 \cos \theta_2} (2mE_1 + 2mE_2) + 4 \sqrt{mE_1 \sqrt{mE_2 \cos \theta_1}} \right]^{1/2}, \tag{2}
\]

where \( \theta_1 \) is the angle between the momentum vectors of the first and second neutrons in the system of the struck nucleus; \( \theta_2 \) is the angle between the resultant of the first two neutrons and the momentum vector of the third neutron; \( \theta_3 \) is the angle between the resultant of all three neutrons and the direction of the incident particle beam; \( E_1, E_2, \) and \( E_3 \) are the kinetic energies of the first, second, and third neutrons respectively in the system of the recoiling nucleus; and \( m \) is the neutron mass, taken as one amu. Energies were calculated in Mev, and masses in amu.

Similarly, for the case of two emitted neutrons, we have

\[
\tan \phi = \frac{\sqrt{1 - \cos^2 \theta_2}}{\cos \theta_2 + \frac{p_a}{p_n}}, \tag{3}
\]

where \( p_n \) is given by

\[
p_n = \left( 2mE_1 + 2mE_2 + 4\sqrt{mE_1 \sqrt{mE_2 \cos \theta_1}} \right)^{1/2}. \tag{4}
\]

All quantities are as defined above, except that here \( \theta_2 \) is the angle between the resultant of the two neutrons and the direction of the incident particle beam.

In order to produce an isotropic neutron distribution, the cosines of the above angles are selected randomly in the range -1 to 1. The neutron energies are selected according to the following scheme:
First, neutrons are selected in a random fashion to fit the distribution
\[ P(E) = E e^{-E/T} \]  \hspace{1cm} (5)
This is done in the case of the first neutron by selecting a neutron energy at random in the range from zero to \( E_a + Q_1 \) by multiplying \( E_a + Q_1 \) by a random number in the range zero to one. \( E_a \) is the energy of the incident particle (a) in the center-of-mass system, and \( Q_1 \) is the Q value for the (a,n) reaction.) The maximum possible value of \( P(E) \) at the particular \( T \) selected is then calculated from Eq. (6),
\[ P(E)_{\text{max}} = M \frac{T}{e} \]  \hspace{1cm} (6)
\( P(E)_{\text{max}} \) is then multiplied by a random number in the range zero to one. If the number thus obtained is greater than \( P(E) \) for the neutron energy selected, this neutron is rejected. The energy spectrum of neutrons surviving this operation is that of Eq. (5). The energies of subsequent neutrons are limited in the selection process to the energy available at that stage of the evaporation.

The next operation is the selection of neutrons from the above spectrum that lead to the reaction under investigation. As an example, let us consider a reaction in which three neutrons are emitted. A flow diagram of this selection process is shown in Fig. 3.

All the above neutron selection operations, as well as the recoil-angle calculations, were performed on an IBM type 701 digital computer. One thousand cases of the reaction under investigation were tabulated at each incident-particle energy studied. The time required for the calculation varied from about 2 minutes at the most favorable energies to much longer times at energies where most of the neutrons were used up by competing reactions.

The following information was recorded for each reaction:
1. The number of recoil events in 1-degree intervals in the laboratory system.
2. The above number modified by solid-angle corrections. (For comparison with experiments, the angular distributions were integrated...
Fig. 3. Flow diagram of Monte Carlo neutron-energy selection scheme. Here $E_a$ is the energy of the incident particle in the center-of-mass system; $Q_1$, $Q_2$, and $Q_3$ are the $Q$ values respectively for the $(\alpha,n)$, $(\alpha,2n)$, and $(\alpha,3n)$ reactions; $E_1 < E_a + Q_1$; $E_2 < E_a + Q_2 - E_1$; and $E_3 < E_a + Q_3 - E_1 - E_2$. 
over intervals corresponding to the range of angles subtended by the rings cut from the catcher foils.)

3. The energies of the neutrons used in the calculation of the recoil angles, tabulated separately for the first, second, and third neutrons emitted, as the number of neutrons per 0.1 Mev interval. (See Fig. 4.)

4. The number and type of other reactions per thousand cases of the reaction under investigation.

The relative frequency of occurrence of the various competing reactions as given by the Monte Carlo calculations was found to be quite sensitive to the value of the parameter T used in the calculation. Although the angular distributions are not much affected by changes in this parameter, an effort was made to use the value of T which gave the best fit to experimental cross-section data. For the system Bi^{209} + He^{4}, it was found that experimental values of the ratio of the (α,2n) and (α,3n) cross sections were matched quite nicely by the Monte Carlo results using a T of 1.4 Mev (Fig. 5). For the system Bi^{209} + H^{2}, however, no single value of T yields cross-section ratios that are in agreement with the experimental data, as may be seen in Fig. 6. By cross-plotting, values of T that fit the experimental data at various energies are obtained (Fig. 7). For this system, values of T read from this graph, and extrapolated values, were used in the Monte Carlo calculations. Calculations were also done based on assumption of a constant T of 1.75 Mev, and these gave angular distributions which agreed with those using values of T from Fig. 7 within the statistics of the 1000 cases tabulated.
Fig. 4. Energy spectra of neutrons leading to the reaction Bi$^{209}$(α,3n)At$^{210}$ (T = 1.40 Mev).
Fig. 5. Ratio of cross sections for the reactions $^{209}\text{Bi}(\alpha,2n)^{211}$Bi and $^{209}\text{Bi}(\alpha,3n)^{210}$At as a function of incident particle energy ($\Delta$ = Monte Carlo, $T = 1.2$; $\bullet$ = Monte Carlo, $T = 1.4$; $\bigcirc$ = Monte Carlo, $T = 1.5$; and $\bigcirc$ = experiment).
Fig. 6. Ratio of cross sections for the reactions $\text{Bi}^{209}(d,2n)\text{Po}^{209}$ and $\text{Bi}^{209}(d,3n)\text{Po}^{208}$ as a function of incident particle energy (Monte Carlo and experimental results).
Fig. 7. T as a function of incident deuteron energy for the system Bi$_{209}$ + H$_2$. 
IV. RESULTS AND CONCLUSIONS

A. Reactions of the System Bi$^{209}$ + He$^4$

1. The ($\alpha$,2n), ($\alpha$,3n), and ($\alpha$,4n) reactions of Bi$^{209}$.

The heavy-recoil angular distributions for the ($\alpha$,2n), ($\alpha$,3n), and ($\alpha$,4n) reactions of Bi$^{209}$ have the general shape shown for the reaction Bi$^{209}$(\$\alpha$,4n)At$^{209}$ in Fig. 8. The angle marked on the abscissa as $\theta_{\text{max}}$ is the maximum angle in the laboratory system to which the recoil may be deflected. This is calculated by assuming that all the neutrons are emitted at the optimum angle and energy for deflecting the nucleus by the maximum amount. All recoils observed at greater angles must therefore be due to scattering caused by the interaction of the recoil atoms with the target material and with the gas molecules in the space between the target and the catcher foil. The deviation of the experimental points in the range of $\theta_{\text{max}}$ from a smooth curve dropping to zero at $\theta_{\text{max}}$ is, then, a measure of the scattering. The variation of the scattering with target thickness was investigated. It was found that for target thicknesses of 1 $\mu$g/cm$^2$ bismuth or less, scattering was negligible at angles in the region of the "half width". The half width (width of the angular distribution at half maximum, measured in degrees) was taken as a measure of the shape of the angular distribution for the purpose of comparison with the Monte Carlo results over a range of incident particle energies.

For the reaction Bi$^{209}$(\$\alpha$,3n)At$^{210}$, the experimental angular distributions and those resulting from the Monte Carlo calculations were found to be in excellent agreement, except for scattering in the region of $\theta_{\text{max}}$, over the entire range of incident particle energies investigated. Experimental and Monte Carlo results for nearly identical incident particle energies are shown for the ($\alpha$,3n) reaction in Fig. 9. For comparison of the experimental and Monte Carlo results at the various energies investigated, the half widths of the angular distributions were plotted as a function of the energy available for neutron evaporation, $E_{\text{cm}} + Q_3$ (Fig. 10).
Fig. 8. Recoil angular distribution for the reaction
\[ \text{Bi}^{209}(\alpha,4n)\text{At}^{209} \quad (E_\alpha + Q = 9.0 \text{ Mev}; \theta_{max} = 26.4^\circ; \]
\[ 0.88 \gamma/cm^2 \text{ Bi}^{209} \).
Fig. 9. Monte Carlo and experimental recoil angular distributions for the reaction Bi\(^{209}\)(\(\alpha,3n\))At\(^{210}\). Standard deviations are shown for the Monte Carlo calculations. The solid curve is drawn through the experimental points (\(E_{\text{cm.}} + Q = 9.7\) Mev for the experiment and 10.0 Mev for the Monte Carlo calculations; (0.88 \(\gamma/\text{cm}^2\) Bi\(^{209}\)).
Fig. 10. Angular distribution half-width as a function of $E_{\text{cm}} + Q$ for the reaction $\text{Bi}^{209}(\alpha,3n)\text{At}^{210}$ ($\bullet$ = experiment, $\bigcirc$ = Monte Carlo, $T = 1.40$ Mev).
The probable error in the half widths of the angular distributions of the \((\alpha,xn)\) reactions depends somewhat on the incident energy and the particular reaction involved. In general, errors are less than \(\pm 1/2\) degree for the experiments and less than \(\pm 1\) degree for the Monte Carlo calculations.

The general features of the curve shown in Fig. 10 are readily explainable on the basis of the model assumed in the Monte Carlo calculations. At the threshold of the reaction, no kinetic energy is available for the outgoing neutrons, so that the neutrons can give no momentum to the recoiling nucleus. Therefore all recoils are found at zero degrees, owing to the forward momentum of the incident particle. As the incident particle energy is increased, the neutrons start contributing to the nuclear recoil, and the half width increases, rapidly at first, then more slowly, until all the neutrons are leaving the nucleus with about the same kinetic energy distribution, as shown in Fig. 4. (The most probable energy under these conditions is about 1 Mev.) The half width then remains nearly constant with increasing energy, the extra energy remaining as excitation energy of the residual nucleus. When the excitation energy of the residual nucleus begins to exceed the binding energy of the next neutron, the \((\alpha,4n)\) reaction begins to compete. Under these conditions, the neutrons must leave with higher kinetic energies in order to give rise to an \((\alpha,3n)\) reaction. This causes the half width to increase again, and this increase will continue ad infinitum, or until another mechanism takes over.

It is interesting that this reaction appears to be explained by an isotropic evaporation model. As has been pointed out by Thomas,\(^54\) Wolfenstein,\(^55\) and Hauser and Feshbach,\(^56\) it can be shown that particle emission from a compound nucleus will be isotropic if the level densities for both the compound and the residual nucleus are sufficiently high, and the levels have a \(2J + 1\) spin dependence over the range of possible \(J\) values. Calculations by Bloch\(^57\) based on the individual-particle model indicate that this dependence is not found at excitation energies less than about 12 Mev. In the reactions studied in this work, excitation energies
much less than this were common. At bombarding energies near the $Q$, the residual nucleus must be left in or near the ground state. However, it is quite possible that the necessary conditions for isotropy are much less stringent than the sufficient conditions described above.

A typical angular distribution for recoils coming from the $\text{Bi}^{209}(\alpha,4n)\text{At}^{209}$ reaction is shown in Fig. 8. Monte Carlo calculations were not done for this reaction because of the complexity of the equations involved, and because the experimental data do not cover a very wide range of incident particle energies. A plot of half width versus energy is shown for this reaction in Fig. 11. This reaction could not be studied at lower energies because the presence of large amounts of $\text{At}^{211}$ alpha activity makes the $\text{At}^{209}$ alpha particles difficult to detect at energies at which the $(\alpha,4n)$ cross section becomes small. The highest energy studied was determined by the maximum energy of the cyclotron.

The general magnitude of half widths for the $(\alpha,4n)$ reaction appears to be about the same as for the $(\alpha,3n)$ reaction at comparable energies. The $(\alpha,4n)$ half widths are slightly greater at low energies. Since there are four particles emitted instead of three, the cancellation of neutron momenta should be more nearly complete, which would give a smaller half width. On the other hand, because the energy is shared among more particles, the absolute value of the total neutron momentum will be greater for the $(\alpha,4n)$ reaction, which tends to increase the half width. It is difficult to say which of these effects would be expected to predominate.

Angular distributions for the $(\alpha,2n)$, $(\alpha,3n)$, and $(\alpha,4n)$ reactions measured at the same energy ($45.5$ Mev) are shown in Fig. 12. The large differences in the half widths are due primarily to the differences in the $Q$ values of the various reactions.

A plot of the half width of the angular distribution versus energy is shown in Fig. 13 for the reaction $\text{Bi}^{209}(\alpha,2n)\text{At}^{211}$. Both Monte Carlo and experimental results are shown. Here it is seen that at values of $E_{\alpha \text{ c.m.}} + Q_2$ greater than about $10$ Mev the experimental half widths lie considerably below those given by the Monte Carlo calculations. Since one could expect the conditions for isotropic neutron
Fig. 11. Angular distribution half-width as a function of $E_{\alpha\text{c.m.}} + Q$ for the reaction Bi$^{209}$ ($\alpha$,ln)At$^{209}$ (experimental).
Fig. 12. Experimental recoil angular distributions for the reactions Bi\textsuperscript{209}(\alpha, 2n)\textsuperscript{211}, Bi\textsuperscript{209}(\alpha, 3n)\textsuperscript{210}, and Bi\textsuperscript{209}(\alpha, 4n)\textsuperscript{209} (E_\alpha = 45.5 MeV).
Fig. 13. Angular distribution half-width as a function of $E_{\text{c.m.}} + Q$ for the reaction Bi$_{209}$($\alpha,2n$)At$_{211}$ ($\bullet$ = experimental; $\circ$ = Monte Carlo).
evaporation from a compound nucleus to be better met at high excitation energies, these results lead one to suspect that a mechanism other than compound-nucleus formation is starting to contribute to the $(\alpha,2n)$ reaction at this energy, which is at the peak of the $(\alpha,2n)$ excitation function measured by Kelly and Segrè. 8

Further evidence that a non-compound-nucleus mechanism begins to contribute to the $(\alpha,2n)$ reaction at energies above 30 Mev is supplied by the results of the recoil range measurements. Results of range measurements for the $(\alpha,2n)$, $(\alpha,3n)$, and $(\alpha,4n)$ reactions at 43.6 Mev are shown in Fig. 14. There is a striking similarity between the range curves of the $(\alpha,3n)$ and $(\alpha,4n)$ reactions. Since the angular distribution studies have indicated that the $(\alpha,3n)$ reaction proceeds by a compound-nucleus mechanism, the similarity in the range curves for these reactions is good evidence that the $(\alpha,4n)$ reaction also proceeds by a compound-nucleus mechanism. The peak of the range curve for either of these reactions may then be interpreted as the range corresponding to recoils having the full momentum of the incident helium ion. The rest of the curve is due to a combination of range straggling and deviations from the most probable recoil momentum caused by imperfect cancellation of the momenta given to the recoils by the outgoing neutrons. As may be seen in Fig. 14, the most probable range for $(\alpha,2n)$ recoils at this energy is considerably less than that for the $(\alpha,3n)$ and $(\alpha,4n)$ reactions. This can only mean that many of the $(\alpha,2n)$ recoils have failed to absorb the full momentum of the incoming helium ion. In other words, a compound nucleus was not formed.

According to the range-energy relation derived by Bohr 58 and Lindhard and Scharff, 59 the ranges of recoils of the mass and energy studied in this work should be proportional to their energies. This type of range-energy relation is also supported by calculations by Harvey 60 based on experimental results of Baulch and Duncan, 61 Leachman and Atterling, 62 and Sikkeland and Ghiorso. 63 If, then, the recoil range is taken as proportional to its energy, this allows one to calculate the energy of the lower range $(\alpha,2n)$ recoils from the ratio of the
Fig. 14. Recoil range curves for the reactions Bi\(^{209}\)(\(\alpha,2n\))\(^{211}\), Bi\(^{209}\)(\(\alpha,3n\))\(^{210}\), and Bi\(^{209}\)(\(\alpha,4n\))\(^{209}\) (\(\bullet\) = \(^{211}\)At, \(\triangle\) = \(^{209}\)At, \(\blacksquare\) = \(^{210}\)At; \(E_\alpha = 43.6\) Mev).
ranges of the $\alpha,2n$ recoils to those of the $\alpha,3n$ or $\alpha,4n$ recoils, since these last two reactions must proceed by a compound-nucleus mechanism on the basis of the results discussed above.

Range curves for the $\alpha,2n$ and $\alpha,4n$ reactions measured at an incident helium ion energy of 46.5 Mev are shown in Fig. 15. The most probable recoil range for the $\alpha,2n$ reaction is seen to be only half that for the $\alpha,4n$ reaction. From this result the $\alpha,2n$ recoil forward momentum is about 0.71 times the $\alpha,4n$ forward momentum. The rest of the forward momentum must be taken by the two emitted neutrons. If we assume all of it is taken by the first neutron, and that this neutron is emitted at 0 degrees, the first neutron would then have a kinetic energy of about 15.6 Mev. In this case the second neutron would have to carry out at least 3 Mev in order not to be followed by the emission of a third neutron. If, on the other hand, the forward momentum were shared equally between the two outgoing neutrons, and the first neutron were emitted near 0 degrees as assumed above, the second neutron would then have to carry out at least 10.8 Mev besides the energy due to its forward momentum. Since this is very unlikely, the first proposed mechanism, or something approximating it, would be expected to predominate. If this is the case, the only momentum available for deviating the path of the recoil from 0 degrees is that due to the second (~3-Mev) neutron. The ratio of this momentum to the forward momentum of the recoiling nucleus is in this case about 0.18. The same ratio for a compound-nucleus mechanism, where both neutrons could contribute to the deflection of the recoil, is about 0.32. Therefore, on the basis of the recoil range measurements, if the $\alpha,2n$ reaction proceeds at these energies by a "knock-on" type mechanism, where the first neutron is ejected with a high kinetic energy at a small angle, one would expect the angular-distribution half widths for this reaction to be less than those predicted by the compound-nucleus (Monte Carlo) calculations. As is seen in Fig. 13, this is indeed the case at energies above about 30 Mev ($E_\alpha \text{c.m.} + Q = \sim 10 \text{ Mev}$). This is further evidence for a "knock-on" mechanism.
Fig. 15. Recoil range curves for the reactions $\text{Bi}^{209}(\alpha,2n)\text{At}^{211}$ and $\text{Bi}^{209}(\alpha,4n)\text{At}^{209}$ ($\bullet = \text{At}^{211}$, $\bigcirc = \text{At}^{209}$; $E_\alpha = 46.5$ Mev).
Results of range measurements at lower energies are shown in Figs. 14, 16, and 17. As may be seen from these figures, the range curve for the \((\alpha,2n)\) reaction gradually approaches that for a compound-nucleus reaction as the incident particle energy is lowered, as would be expected from the Monte Carlo calculations.

2. The Reaction Bi\(^{209}\)(\(\alpha,t\)Po\(^{210}\))

An experimental angular distribution for the heavy recoils coming from this reaction at an incident helium ion energy of 28 Mev is shown in Fig. 18. The limits of error shown are standard deviations calculated from the counting statistics.

From this experiment, the triton angular distribution has been calculated in center-of-mass coordinates on the basis of two assumptions. First, the triton leaves with high enough energy that the residual nucleus is left near the ground state. This seems reasonable, since the \(Q\) for this reaction is \(-14.8\) Mev, leaving after momentum conservation about 12 to 13 Mev for the triton, which is not far above the coulombic potential barrier for bismuth. Second, in the center-of-mass system the tritons are emitted in the great majority of cases at angles less than about 50 degrees. This assumption is necessary because of the double-valued relation between the laboratory and center-of-mass angles of the heavy recoil. (See Ref. 64; \(\gamma = 1.70\) for the ground-state transition.)

Experimental data on triton angular distributions are in agreement with this assumption.\(^{65}\)

The results of the calculation are shown in Fig. 19. It is seen that the tritons are peaked strongly toward small angles. This is consistent with the stripping mechanism that has been proposed for this reaction.\(^{25,65}\)

B. Reactions of the System Bi\(^{209}\) + H\(^2\)

1. The \((d,2n)\) and \((d,3n)\) reactions of Bi\(^{209}\)

Because of the relatively long half lives of the products of the deuteron-induced reactions of Bi\(^{209}\), it was desirable to use somewhat
Fig. 16. Recoil range curves for the reactions

\[ \text{Bi}^{209}(\alpha,2n)\text{At}^{211} \] and \[ \text{Bi}^{209}(\alpha,3n)\text{At}^{210} \] (\( \bullet = \text{At}^{211}, \triangle = \text{At}^{210}, E_\alpha = 38.6 \text{ MeV} \)).
Fig. 17. Recoil range curves for the reactions \( \text{Bi}^{209}(\alpha,2n)\text{At}^{211} \) and \( \text{Bi}^{209}(\alpha,3n)\text{At}^{210} \) (\( \bullet = \text{At}^{211} \), \( \bigcirc = \text{At}^{210} \); \( E_\alpha = 33 \text{ Mev} \)).
Fig. 18. Recoil angular distribution for the reaction Bi\textsuperscript{209}(\alpha,t)Po\textsuperscript{210} (E_\alpha = 28.0 Mev).
Fig. 19. Triton angular distribution in center-of-mass coordinates for the reaction Bi$^{209}$($\alpha$,t)Po$^{210}$ assuming transition to ground state of Po$^{210}$ ($E_\alpha = 28.0$ Mev).
thicker targets in the angular-distribution studies of these reactions than were used in the helium ion bombardments. Because of this factor, and because of the lower momentum of the incident deuterons, it was suspected that scattering might be more extensive than in the case of the helium-ion-induced reactions. In Fig. 20, the half width of the recoil angular distribution of the \((d,3n)\) reaction is shown as a function of target thickness. All these measurements were made at an incident deuteron energy of 23.5 Mev. At target thicknesses of about 6 \(\mu g/cm^2\) bismuth or less, the half width is well within the limits of error (experimental \(\sim \pm 1/2\) degree, Monte Carlo \(\sim \pm 1\) degree) of that for an infinitely thin target. Consequently, targets of about 6 \(\mu g/cm^2\) were used in the angular-distribution studies of the deuteron-induced reactions.

In Fig. 21 are shown experimental and Monte Carlo angular distributions for the \((d,3n)\) reaction at nearly identical incident particle energies. Figure 22 shows the variation of half width with energy for the experiments and Monte Carlo calculations for this reaction. The Monte Carlo calculations are in agreement with the experimental data within the limits of error. It may therefore be concluded that the \((d,3n)\) reaction, like the \((\alpha,3n)\) reaction, proceeds by a compound-nucleus isotropic evaporation mechanism. (See discussion of the \((\alpha,3n)\) reaction.) Apparently the fact that the deuteron is a loosely bound particle of relatively large radius does not prevent its capture as an entity by the nucleus.

The angular distribution of the \((d,2n)\) reaction could be studied only at the energy corresponding to the peak of the excitation function for this reaction, \(\sim 15\) Mev), owing to the long half life of Po\(^{209}\). The angular distribution is shown in Fig. 23. No Monte Carlo calculations were done for this reaction because of the sparsity of experimental data available for comparison. Little can be inferred about the reaction mechanism from the single angular distribution shown in Fig. 23. The curve has much the same shape as that for the \((d,3n)\) reaction at the same energy above the Q.
Fig. 20. Half-width of angular distribution as a function of target thickness for the reaction Bi$^{209}$(d,3n)Po$^{206}$ ($E_d = 23.5$ Mev).
Fig. 21. Monte Carlo and experimental angular distributions for the reaction Bi$^{209}$(d,3n)Po$^{208}$.

(□ = experiment, □ = Monte Carlo (E$_{cm}$ + Q = 11.7 Mev).
Fig. 22. Angular distribution half-width as a function of $E_{dcm} + Q$ for the reaction Bi$_{209}$(d,3n) Po$_{208}$ ($\bullet$ = experiment, $\circ$ = Monte Carlo variable T, $\triangle$ = Monte Carlo T = 1.75).
Fig. 23. Recoil angular distribution for the reaction Bi$^{209}$\,(d,2n)Po$^{209}$ ($E_{\text{cm}} + Q = 10.0$ Mev).
2. The (d,n) and (d,p) reactions of Bi

Recoil angular distributions for the reactions Bi\(^{209}\)(d,n)Po\(^{210}\) and Bi\(^{209}\)(d,p)Bi\(^{210}\) are shown in Figs. 24, 25, and 26. The shapes of the angular distributions for these two reactions differ somewhat, and since the Q values for these two reactions are nearly the same, this may be interpreted as evidence that they are proceeding by different reaction mechanisms.

The angular distributions of some high-energy proton groups coming from the Bi\(^{209}\)(d,p)Bi\(^{210}\) reaction have been measured by Gove \(^66\) and by Wall \(^67\) at a somewhat higher deuteron energy (14 Mev). The angular distribution of these protons exhibits a broad maximum in the region of 60 degrees in the laboratory system. The angular distribution shows no symmetry about 90 degrees and is probably explainable on the basis of a stripping mechanism, \(^68\) although Wall \(^67\) was unable to fit the angular distribution with a Butler \(^69\) type calculation.

Excitation functions for the (d,n) and (d,2n) reactions have been measured by Kelly and Segre. \(^8\) If one makes the assumption that the neutron energy spectrum is of the form \(P(E) = Ee^{-E/T}\), and that the (d,2n) reaction proceeds by a compound-nucleus mechanism at energies below about 15 Mev, it is then possible to calculate from the measured (d,2n) cross sections the cross section of the (d,n) reaction due to the compound-nucleus mechanism. The results of this calculation at several energies in the region studied, together with the corresponding experimental data, are shown below. For the purpose of this calculation, T was taken as 1.75 Mev.

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</table>

The experimental (d,n) cross sections are not in disagreement with those predicted by a compound-nucleus model at energies below about 13 Mev. At higher energies another mechanism must begin to contribute to the reaction.
Fig. 24. Recoil angular distributions for the reactions $\text{Bi}^{209}(d,n)\text{Po}^{210}$ and $\text{Bi}^{209}(d,p)\text{Bi}^{210}$ ($E_d = 9$ MeV).
Fig. 25. Recoil angular distributions for the reactions $\text{Bi}^{209}(d,n)\text{Po}^{210}$ and $\text{Bi}^{209}(d,p)\text{Bi}^{210}$ ($E_d = 10.5$ Mev).
Fig. 26. Recoil angular distributions for the reactions Bi$^{209}$ (d,n)Po$^{210}$ and Bi$^{209}$ (d,p)Bi$^{210}$ ($E_d = 12.5$ Mev).
ACKNOWLEDGMENTS

I wish to thank Professor Glenn T. Seaborg for his interest and encouragement during the performance of this work.

I am greatly indebted to Drs. Bernard G. Harvey and William H. Wade, who developed many of the procedures used, and whose ideas and assistance in the experiments greatly facilitated the progress of this work.

I also wish to thank Dr. José Gonzalez-Vidal for his participation in many lengthy and stimulating arguments.

The assistance of Ernest W. Valyocsik and John Morton with several of the experiments is greatly appreciated, as are the efforts of Robert Freeman in programming the Monte Carlo calculations for the computer.

I am grateful to Dr. Bruce M. Foreman, Glen E. Gordon, Robert J. Silva, Dr. T. Darrah Thomas, and Ernest W. Valyocsik for many helpful discussions.

I wish to thank my wife, Veronica, for her understanding and encouragement during the course of this work.

The cooperation of William Barclay Jones, Peter McWalters, John Wood, Robert Cox, and other members of the 60-inch cyclotron crew is appreciated. Thanks are due Mrs. Patricia Howard for typing the final draft of this report.

I would like to acknowledge the support of the National Science Foundation, which granted me fellowships during the period 1956-1958.

This work was performed under the auspices of the United States Atomic Energy Commission.
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