

## Neutron-Transfer Reactions from the Nitrogen Bombardment of Be, C, O, Na, and Mg<sup>24,25,26</sup>

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Excitation functions were measured for neutron-transfer reactions yielding N<sup>13</sup> from the nitrogen bombardment of Be, C, O, Na, Mg<sup>24</sup>, Mg<sup>25</sup>, and Mg<sup>26</sup>. At the maximum incident energy of 27.5 Mev, the cross section varies from 0.14 mb for oxygen to 2.8 mb for magnesium-25. Nitrogen-13 was separated chemically from the targets and counted as ammonium tetraphenylboron. The chemical method was checked by measuring yields of both reaction products, N<sup>13</sup> and Mg<sup>27</sup>, in the reaction Mg<sup>26</sup>(N<sup>14</sup>,N<sup>13</sup>)Mg<sup>27</sup>. By plotting the transfer cross sections as a function of  $E^* = E_{o.m.} - E_{\text{barrier}} + Q$ , a systematic behavior of all the cross sections is displayed.

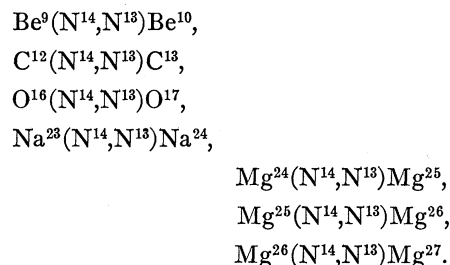
### INTRODUCTION

NUCLEAR reactions induced by heavy ions in light nuclei give evidence of two major reaction mechanisms. One is the formation of a compound nucleus and its subsequent de-excitation by the evaporation of light particles. In activation experiments these reactions are easily identified by the presence of radioactive residual nuclei a few mass units lighter than the compound nucleus.<sup>1,2</sup> The second reaction mechanism, and the one with which the present paper is concerned, results in radioactive residual nuclei one mass unit lighter, or heavier, than either of the two nuclei involved in the reaction. Such residual nuclei can be formed by one of three mechanisms: (1) the transfer of a nucleon from one nucleus to the other in the course of a close collision, (2) the fission of a compound nucleus, and (3) a process in which a nucleon is emitted from one of the colliding nuclei and is not subsequently absorbed. The evaporation of enough particles from a compound nucleus to leave a residual N<sup>13</sup> is always energetically forbidden at bombarding energies available. Process 3 is energetically forbidden in many cases where N<sup>13</sup> is observed after the bombardment of light nuclei by N<sup>14</sup>, and therefore is probably not an important one at incident nitrogen energies of 27 Mev or less. The fission process is also unlikely. It has not been observed in light nuclei by other processes of excitation such as light-particle-induced reactions<sup>3</sup> or photoreactions. The nucleon transfer mechanism then remains the most probable method by which the second kind of heavy-ion nuclear reaction may proceed.

Recently, Breit and Ebel<sup>4</sup> have published theoretical investigations concerning the nucleon transfer process in heavy-ion reactions. It seemed advisable, therefore, to examine systematically such reactions in light nuclei.

High-energy nitrogen ions are particularly well suited for such a study since the transfer of a neutron from the nitrogen to the target nucleus always results in the formation of radioactive N<sup>13</sup>, which can be readily separated chemically and identified by its 10.1-min half-life.

Excitation functions for transfer reactions in boron,<sup>5</sup> nitrogen,<sup>6</sup> and aluminum<sup>7</sup> were measured previously. In the present work the total cross sections were determined as a function of nitrogen energy for the following reactions:



In each case the radioactive N<sup>13</sup> was separated chemically and its yield measured by absolute beta counting. In previous activation experiments<sup>5,8,9</sup> on Be, C, and O, where no chemical separations were performed, no N<sup>13</sup> was found because its low yield was masked by other much more intense activities. Chemical separation was required in the case of sodium and magnesium because of the presence of interfering activities.

Separated isotopes of magnesium were obtained from the Stable Isotopes Division of this Laboratory. Other elements were bombarded in their naturally occurring isotopic abundances.

### EXPERIMENTAL METHOD

Thick targets, described below, were bombarded for 20 minutes with nitrogen ions in the deflected beam of

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<sup>1</sup> Reynolds, Scott, and Zucker, Proc. Natl. Acad. Sci. U. S. 39, 975 (1953).

<sup>2</sup> Halbert, Handley, and Zucker, Phys. Rev. 104, 115 (1956).

<sup>3</sup> B. L. Cohen, Phys. Rev. 102, 453 (1956).

<sup>4</sup> G. Breit and M. E. Ebel, Phys. Rev. 103, 679 (1956); G. Breit and M. E. Ebel, Phys. Rev. 104, 1030 (1956).

<sup>5</sup> Reynolds, Scott, and Zucker, Phys. Rev. 102, 237 (1956).

<sup>6</sup> H. L. Reynolds and A. Zucker, Phys. Rev. 101, 166 (1956).

<sup>7</sup> Webb, Reynolds, and Zucker, Phys. Rev. 102, 749 (1956).

<sup>8</sup> H. L. Reynolds and A. Zucker, Phys. Rev. 100, 226 (1955).

<sup>9</sup> H. L. Reynolds and A. Zucker, Phys. Rev. 101, 166 (1956).

the ORNL 63-inch cyclotron. The incident energy, between 27 and 28 Mev during the course of the experiment, was measured as described elsewhere<sup>5</sup> at the end of nearly every day on which bombardments were made. For bombardments at lower energies thin nickel foils were inserted in the beam. The current was measured with a vibrating reed electrometer; the details of this procedure have been described before.<sup>5</sup>

The chemical method used for the separation of nitrogen was similar for all targets. It consisted essentially of converting the nitrogen to ammonia, distilling the ammonia from an alkaline solution, and precipitating it with sodium tetraphenylboron. The ammonium tetraphenylboron was then thoroughly dried in a counting cup, leaving a smooth white precipitate. The cups were placed under shielded end-window Geiger counters which were calibrated periodically with a standard RaE source.

**Beryllium.**—Disks of beryllium 0.01 in. thick were dissolved, after bombardment in 6*N* HCl containing 1 mg of NH<sub>4</sub><sup>+</sup> as carrier. Then, 5 cc of NaOH were added and the solution was heated; the evolved NH<sub>3</sub> was distilled into a centrifuge tube containing 25 cc of 0.05*N* H<sub>2</sub>SO<sub>4</sub> cooled in an ice bath. Helium was bubbled through the distillation apparatus to speed up the process. NH<sub>4</sub><sup>+</sup> was precipitated by addition of sodium tetraphenylboron solution; the ammonium tetraphenylboron precipitate was centrifuged, washed, and dried in a counting dish.

**Carbon.**—Graphite buttons 0.01 in. thick were dissolved, after bombardment, in an oxidizing solution consisting of fuming sulfuric, phosphoric, and chromic acids. Sodium cyanide equivalent to 1 mg of NH<sub>4</sub><sup>+</sup> was present as carrier. Then, 6*N* NaOH was carefully added to the solution, and NH<sub>3</sub> was separated by distillation as above.

**Oxygen.**—Zinc oxide was used as the target material. The Coulomb barrier of Zn is high enough to prevent nuclear reactions with 27-Mev nitrogen ions. After heating to 350°C to remove carbonates and moisture, ZnO was pressed into  $\frac{3}{4}$ -in. diameter brass molds under a pressure of about 30 tons/in.<sup>2</sup>. In this way a smooth hard target ~0.1 in. thick was produced. After bombardment the target was dissolved in 6*N* NaOH containing 1 mg of NH<sub>4</sub><sup>+</sup> as carrier. Devarda's alloy was added to reduce any oxygen compounds of nitrogen that may have been formed. Ammonia was distilled from solution as outlined above.

**Sodium.**—Sodium bromide targets were made in the same way as the ZnO targets. The bombarded target was dissolved in dilute H<sub>2</sub>SO<sub>4</sub> containing 1 mg of NH<sub>4</sub><sup>+</sup> carrier. Ammonia was distilled as for the other targets.

**Magnesium.**—Isotopically-enriched magnesium was used as target material. The isotopic concentrations of the enriched material are shown in Table I. The separated isotopes in the form of magnesium metal were evaporated onto silver backings. The thickness of magnesium was at least 6 mg/cm<sup>2</sup> in all cases, which

was sufficient to stop the beam as well as all N<sup>13</sup> atoms produced in the target. After bombardment, the target including the silver backing was dropped into a solution of dilute H<sub>2</sub>SO<sub>4</sub> to dissolve the magnesium. The acid solution contained as carrier 1 mg of NH<sub>4</sub><sup>+</sup>. Ammonia was distilled and precipitated as for the other targets.

The chemical separations took from ten to twenty minutes each, depending on the target. Efficiencies of the separations, determined by weighing the dried sample after counting, varied from 40% to 90%. The chemical methods employed here are based on the assumption that N<sup>13</sup>, at the end of its passage through the target, forms a nitride with the metal in the lattice. The possibility of oxygen compounds in the ZnO was also taken into account. In the carbon targets, it was assumed that cyanide is formed. There is, however, no certainty that an unexpected nitrogen compound (i.e., one not converted to ammonia in the chemical processing) may not be formed in these reactions. To check this possibility in magnesium the reaction Mg<sup>26</sup>(N<sup>14</sup>,N<sup>13</sup>)Mg<sup>27</sup> was studied for yields of both the 10.1-min N<sup>13</sup> and the 9.5-min Mg<sup>27</sup>.

The chemical procedure used for the separation of Mg<sup>27</sup> was as follows: The magnesium target was dissolved in 50% acetic acid, hold-back carriers for Na, Si, Cl, Al, K, S, P, and F were added. Boric acid was added to complex F<sup>-</sup>, and magnesium precipitated as the hydroxide with excess 6*N* NaOH. The precipitate was dissolved in a minimum amount of 12*N* HCl, hold-back carriers were added and Mg(OH)<sub>2</sub> precipitated twice more. Magnesium was finally precipitated as magnesium oxalate, MgC<sub>2</sub>O<sub>4</sub>·2H<sub>2</sub>O, washed with glacial acetic acid, transferred to a counting cup, dried, and counted. The time required was about 30 min. The chemical yield was determined, after counting, by titration with disodium dihydrogen ethylenediamine tetraacetate using Eriochrome Black-T as the indicator.

An attempt to perform a similar check on the Na<sup>23</sup>(N<sup>14</sup>,N<sup>13</sup>)Na<sup>24</sup> reaction was unsuccessful because Na<sup>24</sup> is produced copiously by N<sup>14</sup> from small amounts of carbon contamination.

## RESULTS

The nuclear reaction yields as functions of the incident beam energy are shown in Figs. 1 and 2. They were determined from the decay curves after corrections were made for the decay of N<sup>13</sup> during bombardment; corrections were also made for chemical efficiencies and

TABLE I. Percent isotopic concentrations<sup>a</sup> of enriched magnesium targets.

	Mg <sup>24</sup>	Mg <sup>25</sup>	Mg <sup>26</sup>
Enriched Mg <sup>24</sup>	99.59%	0.301%	0.14%
Enriched Mg <sup>25</sup>	1.5%	97.5%	1.0%
Enriched Mg <sup>26</sup>	0.6%	0.4%	98.99%

<sup>a</sup> The isotopic concentrations of the enriched magnesium were supplied by the Stable Isotopes Division, Oak Ridge National Laboratory.

backscattering. In calculating the yields from  $Mg^{26}$  targets, the  $N^{13}$  produced from the 0.4%  $Mg^{25}$  present in the target was taken into account. This correction lowered the observed yield by as much as 20% at some energies. Similar corrections for the other magnesium targets were negligible.

The relative errors in any cross-section curve are about  $\pm 15\%$ , while the absolute errors in the cross section are about  $\pm 20\%$  for the high-yield reactions, and about  $\pm 30\%$  for C, O, and  $Mg^{26}$  targets. The chief source of error in the yield measurements lay in the calibration of the Geiger counters. Other contributing sources of error may be attributed to uneven deposition of ammonium tetraphenylboron in the counting dish, the uncertainties in the backscattering correction, source thickness, and small errors in measuring time and in calibration of the beam-current integrator. For low-yield points the errors due to counting statistics become appreciable (about 15% for yields near  $10^{-10}$ ).

The yields of 9.5-min  $Mg^{27}$  from the reaction  $Mg^{26}(N^{14},N^{13})Mg^{27}$  are shown as crosses in Fig. 2. These yields agree with the  $N^{13}$  yields from the same reaction and show that, at least for magnesium, the  $N^{13}$  forms a metal nitride when it comes to rest, or that some other compound is formed which acts like the nitride insofar as the evolution of ammonia is concerned.  $Mg^{27}$  yields obtained from natural magnesium and enriched magnesium 26 targets agree with one another as well as with the  $N^{13}$  yield after the correction for isotopic abundance is applied. This shows that the double-neutron-transfer cross section for the reaction

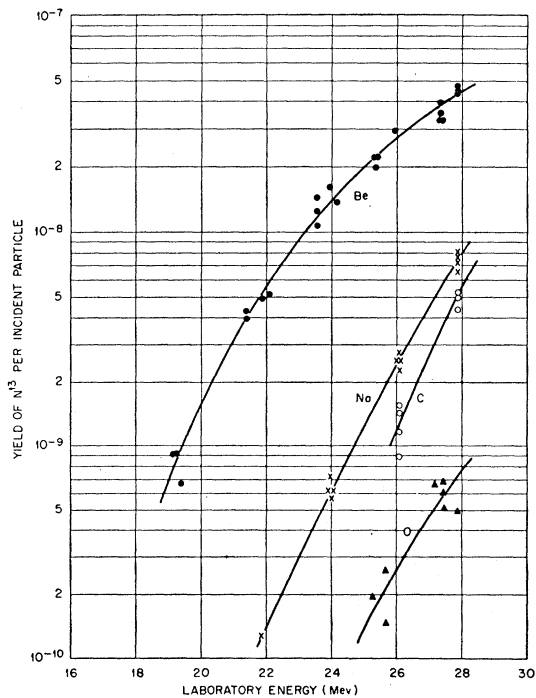


FIG. 1. Yield of  $N^{13}$  per incident  $N^{14}$  ion from beryllium, carbon, oxygen, and sodium.

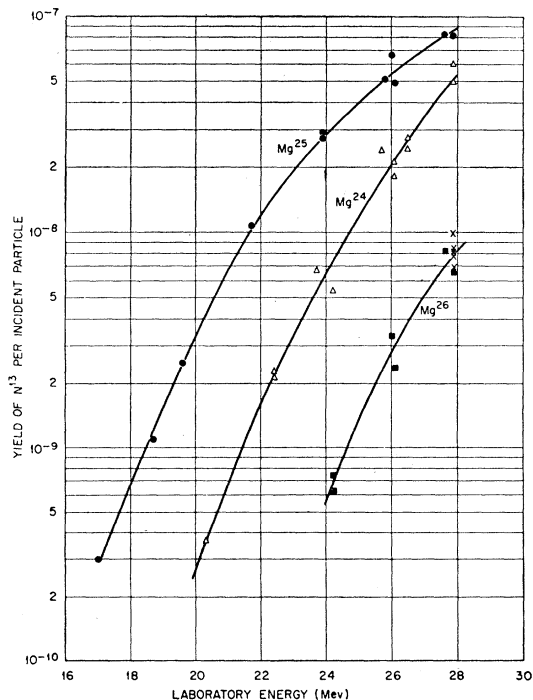


FIG. 2. Yield of  $N^{13}$  per incident  $N^{14}$  ion from the separated magnesium isotopes. The crosses are the yield of  $Mg^{27}$  from the reaction  $Mg^{26}(N^{14},N^{13})Mg^{27}$ .

$Mg^{25}(N^{14},N^{12})Mg^{27}$  is small compared to the single-neutron-transfer cross section. It also gives assurance that the 10-min activity assumed to be due to  $Mg^{27}$  was certainly not  $N^{13}$ .

The effect of impurities in the targets was generally not important. Such common impurities as carbon and oxygen have very low yields of  $N^{13}$  and could therefore not seriously affect the results in targets where carbon or oxygen might be present to the order of even as much as a few percent. The presence of nitrogen impurity was more serious, since (a) the yield of  $N^{13}$  from  $N^{14}$  is relatively high, and (b) the impurity nitrogen, contributing to the tetraphenylboron precipitate, introduces an error into the measurement of the chemical separation efficiency. Since the total amount of carrier added was the equivalent of 1 mg of  $NH_4^+$ , as little as one part in ten thousand of nitrogen in a target weighing one gram would create a serious error in the calculation of chemical yields. The nitrogen content of targets was therefore determined chemically and found to be not more than a few hundredths of a percent in the worst case. This did not affect the reaction yields, but it did have to be taken into account in calculating chemical yields.

Smooth lines drawn through the yield points were differentiated to obtain the cross sections as a function of incident energy shown in Figs. 3 and 4. To perform this calculation it was assumed that the range of nitrogen in any material may be calculated from the

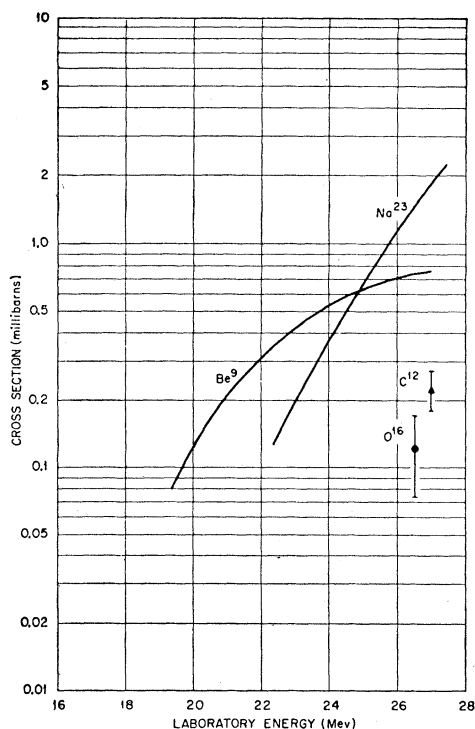


FIG. 3. Total cross sections as a function of incident energy for nitrogen-induced neutron transfer reactions yielding  $N^{13}$  from beryllium, carbon, oxygen, and sodium.

known range of nitrogen in nickel and the relative stopping powers of nickel and of the material in question for protons of the same velocity as the nitrogen ions. This assumption had previously been verified for the range of nitrogen in aluminum.<sup>7</sup>

The presence of  $C^{13}$  in the carbon, and  $O^{17}$  and  $O^{18}$  in the oxygen was also considered. If it is assumed that the transfer cross sections for these isotopes behave normally (see discussion and Fig. 5), then the estimated contribution of  $O^{17}$  and  $O^{18}$  amounts to only about 0.003 mb and can be neglected. However, the  $C^{13}$  contribution is estimated to be of the order of 0.03 mb. The cross section plotted in Figs. 4 and 5 incorporates this correction.

#### DISCUSSION

From the earliest experiments with nitrogen ions it has been observed that transfer cross sections are very  $Q$ -dependent. Even if enough energy is available in the center-of-mass system for the reaction to take place, a  $Q$  more negative than one or two Mev usually means that the cross section is very low.<sup>1</sup> This was made quite apparent when it was found that the transfer cross section for aluminum<sup>6</sup> was 1/40 that for nitrogen and boron. At that time it was thought that three factors might be important in lowering the aluminum transfer cross section: (1) the Coulomb barrier, (2) the  $Q$  of the reaction, and (3) that the stripped neutron makes a

transition from a  $p$  shell in nitrogen to a  $d$  shell in aluminum. To conserve parity, this transition necessitates a change in the relative angular momentum of the two nuclei; this may make the aluminum transfer reaction less likely than, say, the nitrogen or boron reaction where no angular momentum change is required.<sup>10</sup>

In order to test the validity of (3) the measurements on separated magnesium were undertaken. It can be seen from the results that the cross sections for the three magnesium isotopes are very different although conditions (1) and (3) are essentially unchanged. It is therefore apparent that the  $Q$  value plays an important role in the cross section of transfer reactions.

To make this discussion more comprehensive the data previously obtained for the neutron transfer reactions in boron, nitrogen, and aluminum are included. Table II lists the observed reactions, the maximum incident kinetic energy available in the center-of-mass system, ( $E_{c.m.}$ ), ground-state  $Q$  value, and the Coulomb barrier. Coulomb barriers were calculated from the relation

$$E_{barr.} = Z_1 Z_2 e^2 / (A_1^{1/3} + A_2^{1/3}) r_0,$$

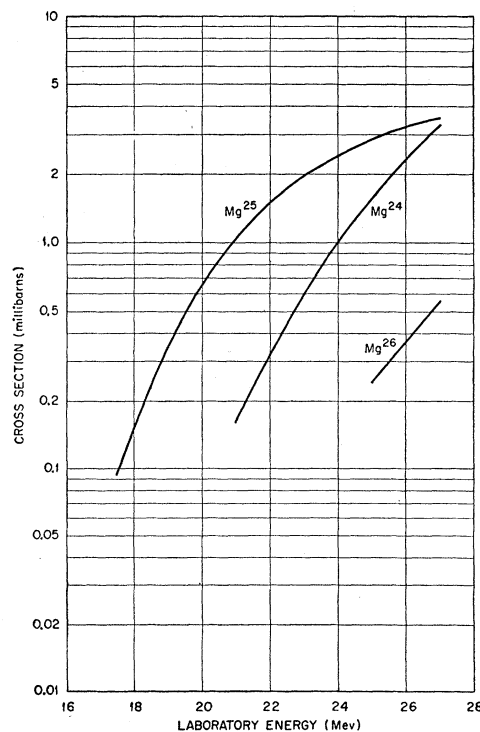


FIG. 4. Total cross sections as a function of incident energy for nitrogen-induced neutron transfer reactions yielding  $N^{13}$  from the separated magnesium isotopes.

<sup>10</sup> This suggestion was originally made by G. Breit (private communication). However, since the work of Breit and Ebel (reference 4) has shown that virtual state formation in either nucleus is important in transfer reactions, parity conservation may be considered as relatively unimportant so far as the total cross sections are concerned.

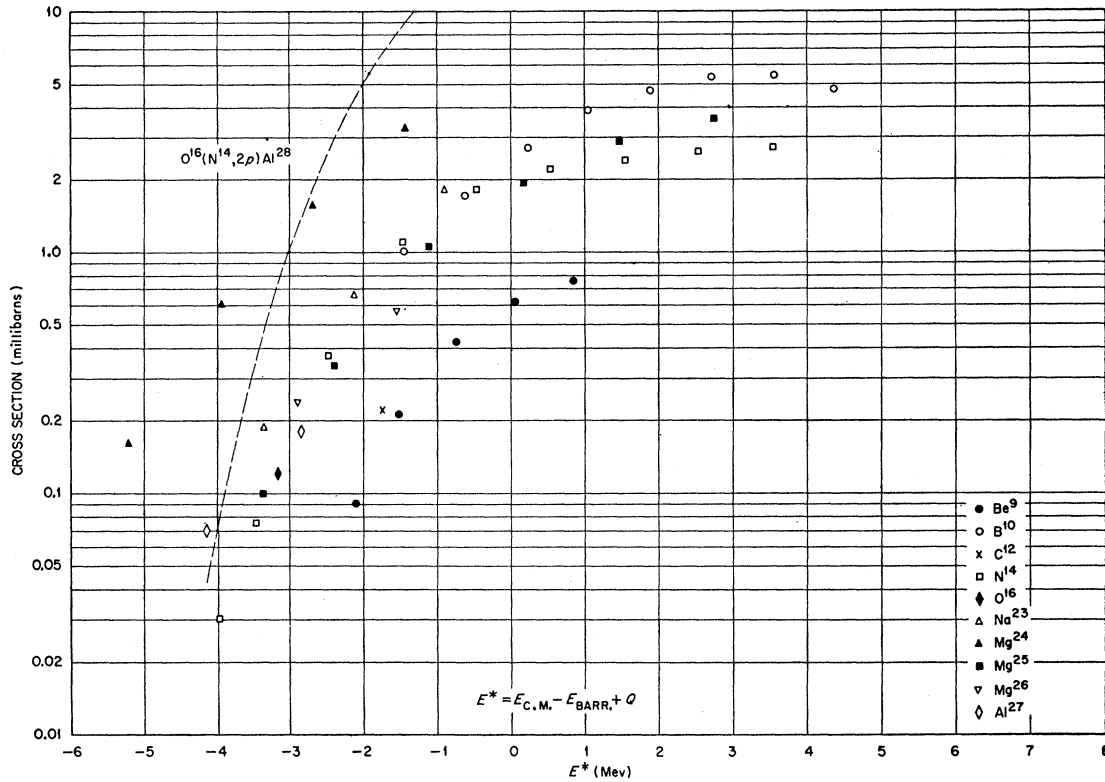


FIG. 5. Total neutron transfer cross sections for ten light elements plotted as a function of  $E^* = E_{c.m.} - E_{barr.} + Q$ , where  $E_{c.m.}$  is the maximum incident kinetic energy available in the center-of-mass system and  $E_{barr.}$  is the Coulomb barrier energy. The dashed line is the excitation function for an evaporation type reaction,  $O^{16}(N^{14}, 2p)Al^{28}$ .

with  $r_0 = 1.5 \times 10^{-3}$  cm used throughout. Also listed in Table II is a quantity  $E^*$  defined by

$$E^* = E_{c.m.} - E_{barr.} + Q.$$

Physically  $E^*$  is the kinetic energy in the system at the moment of contact of the two nuclei just after the neutron has left the nitrogen and attached itself to the target nucleus. Obviously this is a classical picture and takes into account neither the quantum mechanical description of the orbits, nor the details of the transfer mechanism. It is clear that the transfer mechanism

TABLE II. Neutron transfer reactions from nitrogen to various light elements at maximum bombarding energy. All energies are in Mev.

Reaction	$E_{com}$	$E_{barr.}$	$Q^a$	$E^*$
$Be^9(N^{14}, N^{13})Be^{10}$	10.57	5.99	-3.73	+0.84
$B^{10}(N^{14}, N^{13})B^{11}$	10.83	7.36	+0.91	+4.38
$C^{12}(N^{14}, N^{13})C^{13}$	12.46	8.58	-5.61	-1.73
$N^{14}(N^{14}, N^{13})N^{15}$	13.00	9.76	+0.29	+3.53
$O^{16}(N^{14}, N^{13})O^{17}$	14.13	10.91	-6.41	-3.19
$Na^{23}(N^{14}, N^{13})Na^{24}$	16.78	14.07	-3.59	-0.88
$Mg^{24}(N^{14}, N^{13})Mg^{25}$	17.05	15.23	-3.22	-1.40
$Mg^{25}(N^{14}, N^{13})Mg^{26}$	17.31	15.12	+0.57	+2.76
$Mg^{26}(N^{14}, N^{13})Mg^{27}$	17.55	15.01	-4.11	-1.57
$Al^{27}(N^{14}, N^{13})Al^{28}$	16.13	16.15	-2.83	-2.85

<sup>a</sup> The  $Q$  values were calculated from the tables given by A. H. Wapstra, *Physica* 21, 367 (1955).

plays an important role in the cross sections, as has been shown theoretically by Breit and Ebel.<sup>4</sup>

Figure 5 shows a plot of the cross sections for neutron transfer for ten different targets as a function of  $E^*$ . The nitrogen-nitrogen transfer cross section has been divided by two for this plot since the measured cross section includes neutron transfers from either the target nucleus or the incident nucleus.

It can be seen from Fig. 5 that most of the reaction cross sections follow a smooth curve. The most obvious exceptions to this trend are  $Mg^{24}$  which seems to be displaced to the left and  $Be^9$  which is too low. The latter may be understood if details of the nuclear structure are considered. For example, since the odd neutron in  $Be^9$  is bound very loosely, formation of  $Be^{10}$  requires a major change in the wave function of this neutron. The overlap of the initial and final wave functions is relatively small, making the transition unlikely.<sup>11</sup> Other explanations, also based on the loose structure of  $Be^9$ , are possible.

It must be pointed out that a plot such as the one in Fig. 5 can show only general reaction trends since the details of the transfer mechanism have not been taken into account. Figure 5 does, however, demonstrate the importance of the  $Q$  value for transfer cross sections.

<sup>11</sup> A. M. Lane (private communication).

One should point out that even the maverick  $Mg^{24}$  and  $Be^9$  exhibit excitation functions very similar in shape to the general trend. This becomes even more obvious when the transfer reaction excitation functions are compared to a typical excitation function for an evaporation reaction such as  $O^{16}(N^{14}, 2p)Al^{28}$ . It can be seen that the slopes for  $E^* < 0$  are different, and for  $E^* > 0$  the cross section for  $O^{16}(N^{14}, 2p)Al^{28}$  keeps rising

to well above 100 mb at  $E^* = 2.35$  Mev, whereas the transfer cross sections level off around 3 mb.

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## Inelastic Scattering of 40.2-Mev Alpha Particles\*

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The differential cross section  $d^2\sigma/dE d\Omega$  has been measured at 25, 45, 90, 135, and 150 degrees in the laboratory system for the inelastic scattering of 40.2-Mev alpha particles from Al, V, Cu, Nb, Ag, Ta, and Th. Spectra obtained at 135 and 150 degrees have been analyzed in terms of the statistical theory of nuclear reactions. The differential cross section measured at smaller angles is incompatible with the statistical theory of nuclear reactions. A group of alpha particles with energy well below the Coulomb barrier is observed in the spectra of Ta and Th at large angles.

### INTRODUCTION

IN the past few years the experimental evidence on the interactions of heavy particles with energies of about 40 Mev has been studied by several experimenters.<sup>1</sup> The relevant conclusions arrived at as far as the present experiment is concerned are that (1) the nucleus is at its maximum opacity, and consequently the mean free path is at a minimum; (2) the level densities of nuclei increase exponentially with excitation energy; (3) the importance of noncompound nucleus processes has been observed (in some reactions the noncompound nucleus cross section is an order of magnitude larger than the cross section for compound nucleus processes), and (4) in the optical-model treatments of elastic scattering of protons<sup>2</sup> and alpha particles<sup>3,4</sup> it is necessary to introduce a nuclear potential with a diffuse surface.

These conclusions can be given an additional test by the investigation of the interactions of energetic alpha particles with nuclei. According to these conclusions, when an energetic alpha particle strikes the dense, central part of a heavy nucleus, it imparts its

energy and momentum to the nucleons of the nucleus in a series of collisions since the mean free path is considerably shorter than the radius of the nucleus.<sup>4</sup> According to the statistical theory of nuclear reactions, the compound nucleus which results from the interaction emits particles isotropically.<sup>5</sup>

On the other hand, according to the conclusions stated above, the mean free path is long enough so that collisions in the "skin" of the nucleus can occur in which only one nucleon of the nucleus is involved. Collisions of this type would result in an angular distribution peaked strongly in the forward direction.<sup>6,12</sup> Consequently compound nucleus processes predominate in the backward hemisphere, since surface interactions should have small cross sections in the backward hemisphere. The  $(\alpha, p)$  experiment at 40 Mev<sup>7</sup> gives weight to the idea that statistical processes predominate in the backward hemisphere. Consequently a method is available for measuring energy-level densities, namely, by measuring energy spectra in the backward hemisphere.

In the present investigation of inelastic alpha-particle events an independent check is possible on the conclusions reached from the analysis of the 40.2-Mev  $(\alpha, p)$  experiment; namely, that the backward scattering is mainly due to statistical processes.

In addition the inelastic scattering of alpha particles into the forward hemisphere and the cross section for

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<sup>1</sup> "Statistical Aspects of the Nucleus," Brookhaven National Laboratory Report BNL-331(C-21), 1955 (unpublished).

<sup>2</sup> Melkanoff, Nadrick, Saxon, and Woods, University of California at Los Angeles Report UCLA-7-12-55 (unpublished).

<sup>3</sup> Cheston, Glassgold, Stein, Schuldt, and Erickson, *Bull. Am. Phys. Soc. Ser. II*, **1**, 339 (1956).

<sup>4</sup> Igo, Thaler, and Hill, *Bull. Am. Phys. Soc. Ser. II*, **1**, 384 (1956).

<sup>5</sup> See for instance J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

<sup>6</sup> R. M. Eisberg and G. Igo, *Phys. Rev.* **93**, 103 (1954).

<sup>7</sup> Eisberg, Igo, and Wegner, *Phys. Rev.* **100**, 1309 (1955).