

anisotropic than fission at higher excitation energy. These two effects may be entirely unconnected and yet, in any bombardment, they would be responsible for an observable correlation between mass and angular distributions. The fissions taking place before much neutron emission would be rather symmetric and isotropic; those occurring later would be asymmetric and anisotropic. Here again a semiquantitative analysis shows that it is not impossible that the observed correlations are entirely due to this "coupling" through neutron evaporation. §

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Reactions of Uranium-238 with Carbon Ions*

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The excitation functions for some reactions of U^{238} with monoenergetic C^{12} ions have been measured by use of the stacked-foil technique. The $(C,4n)$ and $(C,6n)$ reactions were found to occur through the formation of a compound nucleus followed by neutron evaporation. The results were consistent with calculations made by a modified Jackson-type treatment. Application of the information from the $U^{238}(C,xn)$ reactions to the calculation of cross sections for the $Pu^{242}(C,4n)Fm^{250}$ reaction was found to give agreement with experimental results.

The $(C,\alpha 4n)$ reaction probably proceeds mainly by a stripping mechanism, but there is also an indication of evaporation of alpha particles from a compound system.

INTRODUCTION

MOST of the quantitative information on nuclear reactions in the heavy-element region has been confined to investigations with helium ions or lighter particles because of the difficulties of obtaining intense monoenergetic beams of heavier ions. Heavy ions such as carbon, nitrogen, oxygen, and neon have been accelerated in cyclotrons, but usually with relatively low intensities and with broad energy spectra so that quantitative interpretation of the experimental results is difficult.¹⁻⁷ However, more recently, investigations have been made in Russia of the dependences of the spallation cross sections of gold bombarded with monoenergetic nitrogen ions using the 150-cm cyclotron of the ANSSSR.⁸ The cross-section curves exhibited sharp

maxima consistent with the theory of evaporation processes. Another group at the same location studied the fission cross sections of ytterbium, rhenium, gold, bismuth, U^{235} , and U^{238} as functions of energy,⁹ using the monoenergetic nitrogen ions.

The use of a linear accelerator for obtaining beams of monoenergetic heavy ions has many advantages. It is readily adaptable to the acceleration of a wide range of different ions. The linear accelerator can be changed rapidly to accelerate the desired particles, the external beams are well focused and of high intensities, and the particles are of well-defined energies. Such linear accelerators have been constructed at Berkeley^{10,11} and at Yale University. These accelerators produce ions with energies of 10 Mev per nucleon.

The heavy-element region is particularly interesting for the study of the reaction mechanisms, since fission competition has a large influence on the spallation cross sections.¹² A knowledge of the excitation functions with

* This work was performed under the auspices of the U. S. Atomic Energy Commission.

¹ L. W. Alvarez, *Phys. Rev.* **58**, 192 (1940).

² York, Hildebrand, Putnam, and Hamilton, *Phys. Rev.* **70**, 446 (1946).

³ Ghiorso, Thompson, Street, and Seaborg, *Phys. Rev.* **81**, 154 (1951).

⁴ Rossi, Jones, Hollander, and Hamilton, *Phys. Rev.* **93**, 256 (1954).

⁵ Ghiorso, Rossi, Harvey, and Thompson, *Phys. Rev.* **93**, 257 (1954).

⁶ Fremlin, Glover, and Milsted, *J. Inorg. Nuclear Chem.* **2**, 263 (1956).

⁷ H. Atterling, *Arkiv Fysik* **7**, 503 (1954).

⁸ Baraboshkin, Karamian, and Flerov, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **32**, 1294 (1957) [translation: *Soviet Phys. JETP* **5**, 1055 (1957)].

⁹ Druin, Polikanov, and Flerov, *J. Exptl. Theoret. Phys. (U.S.S.R.)* **32**, 1298 (1957) [translation: *Soviet Phys. JETP* **5**, 1059 (1957)].

¹⁰ Beringer, Gluckstern, Malkin, Hubbard, Smith, and Van Atta, University of California Radiation Laboratory Report UCRL-2796, November, 1954 (unpublished).

¹¹ Gerdinand Voelker, University of California Radiation Laboratory Report UCRL-3777, August, 1957 (unpublished).

¹² Glass, Carr, Cobble, and Seaborg, *Phys. Rev.* **104**, 434 (1956).

heavy ions is also of interest in the production of new heavy isotopes and new elements.¹³

It was convenient to begin the general investigation of heavy-ion reactions in the heavy-element region with bombardments of U^{238} with C^{12} ions. U^{238} is available in large amounts and has a low specific activity, and is therefore readily adaptable to the stacked-foil technique. Most of the spallation products are well known; they decay by alpha-particle emission and have favorable half-lives.

EXPERIMENTAL

Bombardments

The stacked foils were made by vaporizing UF_4 onto nickel foils of 2.3 mg/cm^2 thickness. U^{238} (99.3%) was used and its thickness was approximately 1 mg/cm^2 . In front of the foils was an aluminum absorber of 14.1 mg/cm^2 thickness, which reduced the energy of the carbon ions from 120 to 102 Mev. A collimator 0.91 by 0.50 cm was used so that the total beam measured by the Faraday cup would pass through the UF_4 . For each foil the UF_4 layer was facing the beam; therefore all spallation recoils were stopped in either the UF_4 layer or the nickel backing. The energies of the C ions in the different foils were evaluated from calculated range-energy curves for carbon in uranium, nickel, and fluorine.¹⁴ Eighteen foils were used and the energy in the last one was 45 Mev. The intensity of the beam was assumed to be constant through the foils, and equal to that measured by the Faraday cup. The carbon ions were assumed to be fully stripped after passing through the foils. The pulses were of 2-millisecond duration and the time between the pulses was 0.5 sec, giving a duty cycle of 4×10^{-3} . The measured beam varied between 2 and 20 millimicroamperes with an average intensity of $11 \text{ m}\mu\text{a per cm}^2$. Since the time of these experiments several improvements have been made which have increased the beam intensity to approximately one microampere.

Chemistry

The different foils were dissolved in concentrated HCl containing a trace of H_2O_2 , H_3BO_3 , and HNO_3 . The transplutonium elements in 6M HCl were separated from uranium by sorbing the latter on a column packed with Dowex-1 anion-exchange resin, and from nickel by elution with 6M HCl from a column packed with Dowex-50 cation-exchange resin.^{15,16} The higher actinides are eluted after nickel and before thorium and actinium. The transplutonium-element fraction was

finally electroplated on platinum plates¹⁷ and was subjected to alpha-particle pulse-height analysis. The different nuclides were identified by their alpha energies and decay properties. In each case the chemical yield was determined by using Am^{241} yield tracer in the solution.

Results

The cross sections of the spallation products Cf^{246} , Cf^{244} , and Cm^{242} as a function of the C^{12} ion energy in the laboratory system are given in Fig. 1. The errors given are statistical errors. The yield of Cm^{242} is corrected for the growth of Cm^{242} from the decay of Cf^{246} .

The spallation product Cm^{240} (27-day half-life) was also observed, but with large errors in measurement resulting from the low levels of activities. We have, therefore, not included the low-energy part of its curve.

Products from the reaction $(C,4n)$ are observed 2

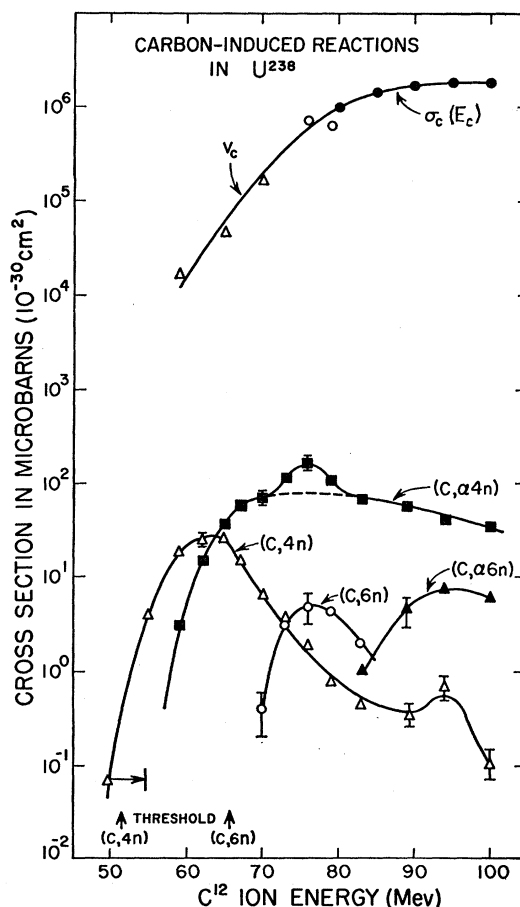


FIG. 1. C^{12} induced reactions in U^{238} . Legend for $\sigma_c(E_c)$ curve: Δ calculated from the $(C,4n)$ cross section; \circ calculated from the $(C,6n)$ cross section; \bullet calculated from the formula $\sigma_c(E_c) = \pi(R_u + R_c)^2(1 - V_c/E_c)$.

¹³ S. G. Thompson, *Svensk Kem. Tidskr.* **69**, 357 (1957).

¹⁴ Rosemary Barrett (private communication, 1957).

¹⁵ Diamond, Street, and Seaborg, *J. Am. Chem. Soc.* **76**, 1461 (1954).

¹⁶ Thompson, Harvey, Choppin, and Seaborg, *J. Am. Chem. Soc.* **76**, 6229 (1954).

¹⁷ Harvey, Chatham-Strode, Ghiorsio, Choppin, and Thompson, *Phys. Rev.* **104**, 1315 (1956).

Mev below the threshold. This may arise from the following sources:

1. Straggling of the carbon ions in the foils.
2. Imperfections in the foils.
3. Errors in measurements of the thickness of the foils.
4. Uncertainties in the maximum energies of the ions accelerated by the linear accelerator.
5. Errors in the range-energy curves.

Straggling in the foils will increase the energy distribution by 0.3 Mev at the $(C,4n)$ threshold.¹⁸ Both 1 and 2 should make the observed peak of the excitation function curve for the $(C,4n)$ reaction broader, whereas it is actually at least as narrow as that calculated from Jackson's formula. This is also demonstrated by the curve for the reaction $Pu^{242}(C,4n)Fm^{250}$ (Fig. 2). Item 3 should not introduce larger errors than 0.5 Mev and item 4 negligible errors in this energy range.

These suggest therefore possible systematic errors in the range-energy curves. These have been evaluated from data for ions not heavier than helium,¹⁹ because accurate range-energy measurements have not been made for heavy ions. The combined error from all sources is believed to be less than 5 Mev.

The point on the $(C,4n)$ cross-section curve corresponding to a carbon-ion energy of 94 Mev seems high by a factor of three, a discrepancy which cannot be explained by statistical errors. This might be an indication of the presence of a lower-energy component in the beam.

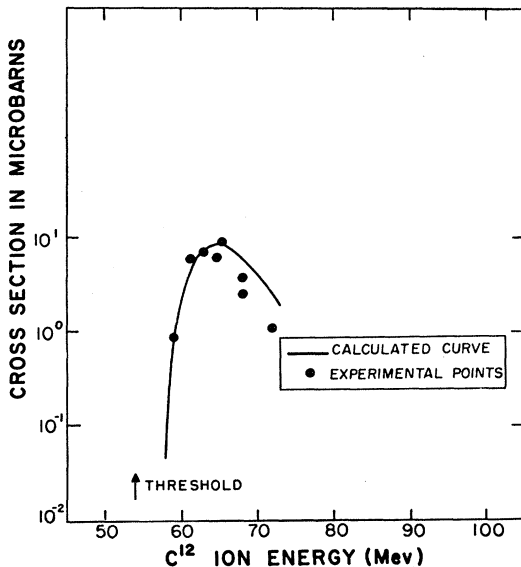


FIG. 2. Excitation function for the reaction of $Pu^{242}(C^{12},4n)Fm^{250}$.

¹⁸ R. Gluckstern, Berkeley Engineering Notes 7317-30 CV-11, December, 1954 (unpublished).

¹⁹ Aron, Hoffman, and Williams, Atomic Energy Commission Report AECU-663, October, 1949 (unpublished).

DISCUSSION

A. (C,xn) Reactions

The $(C,4n)$ and $(C,6n)$ curves are characterized by sharp peaks indicating compound-nucleus formation followed by neutron evaporation. If this interpretation is correct the cross section, $\sigma_{(C,xn)}$, may be expressed according to a modified Jackson formula²⁰:

$$\sigma_{(C,xn)} = \sigma_c(E_C) G_1 G_2 \cdots G_x P_{xn}(E^*) \quad (1)$$

Where $\sigma_c(E_C)$ is the cross section for the formation of the compound nucleus Cf^{250} at the bombarding energy E_C of the C^{12} ions.

$$G_i = (\Gamma_n/\Gamma_i) \simeq [\Gamma_n/(\Gamma_n + \Gamma_f)]_i$$

is the branching ratio for the emission of the i th neutron in an evaporation process. The product $(\bar{G})^x = (G_1 G_2 \cdots G_x)$ therefore gives the fraction of the initially formed compound nuclei which survive fission in x successive evaporations of neutrons.

$$P_{xn}(E^*) = I(\Delta_x, 2x-3) - I(\Delta_{x+1}, 2x-1)$$

is the probability for evaporating exactly x neutrons at the excitation energy E^* , where $I(z,n)$ is Pearson's incomplete gamma function.

$$\Delta_x = (E^* - \sum^x B_i/T),$$

where B_i is the binding energy of the i th neutron²¹ and T is the nuclear temperature (assumed to be constant through the evaporation process).

$$\Delta_{x+1} = (E^* - \sum^{x+1} B_i)/T \quad \text{or} \quad (E^* - \sum^x B_i - E_{th})/T,$$

when $E_{th} < B_{x+1}$,

where E_{th} is the activation energy for fission.²²⁻²⁴ The nuclear temperature for fission is assumed to be equal to that for neutron evaporation.

The $\sigma_c(E_C)$ can be calculated for carbon ions of energies (E_C) greater than 1.2 times the Coulomb barrier (V_C) by use of the formula²⁵

$$\sigma_c(E_C) = \pi(R_U + R_C)^2 (1 - V_C/E_C), \quad (2)$$

where $R = r_0 A^{1/3}$, V_C = Coulomb barrier, and E_C = energy of the carbon ions.

At lower energies the calculations are complicated and uncertain. Instead we shall use formula (1) to evaluate $\sigma_c(E_C)$.

We shall now make the following assumptions:

(a) For the average values of $\bar{G} \equiv (G_1 G_2 \cdots G_x)^{1/x}$, we use those calculated from helium-induced reactions on

²⁰ J. D. Jackson, Can. J. Phys. **34**, 767 (1956).

²¹ Glass, Thompson, and Seaborg, J. Inorg. Nuclear Chem. **1**, 3 (1955).

²² W. J. Swiatecki, Phys. Rev. **101**, 651 (1956).

²³ J. D. Jackson, Chalk River Symposium on Fission, May, 1956, CRP-642A (unpublished), paper B2.

²⁴ R. Vandenbosch and G. T. Seaborg, Phys. Rev. **110**, 507 (1958).

²⁵ J. B. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

nuclei, which give the same compound nuclei as those for the heavy ions. The \bar{G} values are those obtained for $(\alpha, 4n)$ reactions which also proceed mainly through compound-nucleus formation and neutron evaporation. In this case the process following the compound-nucleus formation is independent of the mode of formation and the \bar{G} values should be the same for helium-ion and carbon-ion reactions. When experimental data are unavailable, an estimate of the \bar{G} may be obtained from the systematics for $(\alpha, 4n)$ reactions compiled by Vandenbosch and Seaborg.²⁴ In this way we obtain the values $(\bar{G})^4 = 1.6 \times 10^{-3}$ and $(\bar{G})^6 = 1.0 \times 10^{-5}$. We shall also assume \bar{G} to be independent of the excitation energy,¹² $\partial \bar{G} / \partial E^* = 0$, in the energy range involved.

(b) Direct processes may be neglected, since the probability should be small that a heavy ion of energy less than 10 Mev per nucleon could knock out four neutrons.

The best fit to a smooth curve for σ_c was obtained by using the parameter $r_0 = 1.5$ fermis (1 fermi $\equiv 10^{-13}$ cm). This is the same as that found to give the best fit for helium-induced reactions on heavy nuclei.²⁶ For nitrogen-induced reactions a parameter $r_0 = 1.55$ fermis has given a good fit.⁹ For the average nuclear temperature, T , we have to use 0.90 Mev at the peak for the $(C, 4n)$, and we have to increase it to 1.06 Mev at the peak for the $(C, 6n)$ reaction in order to obtain a good fit. The results of these calculations are expressed by the curve for $\sigma_c(E_C)$ in Fig. 1.

The increase of the nuclear temperature with excitation energy is in accordance with what might be expected from simple theory. This indicates that the assumption of constant nuclear temperature implicit in Jackson's treatment is not rigorously justified. The value for the nuclear temperature is lower than that found in helium-induced reactions.²⁶ The nuclear temperature obtained in this work, however, may be too low by as much as 0.6 Mev owing to systematic errors in the energies of the carbon ions. A very much higher nuclear temperature must be assumed in interpreting the results of Baraboschkin *et al.* on the reactions of gold with nitrogen ions.⁸ It might well be that the discrepancy is due to uncertainties in the energies of the heavy ions.

The errors in $\sigma_c(E_C)$ given in Fig. 1 now are of the order of the errors for the (\bar{G}) factors, which might be as high as 50%.

A very large fraction of the reaction products following compound-nucleus formation in very heavy isotopes consists of fission products. It should therefore be possible to obtain a good estimate of $\sigma_c(E_C)$ by measuring the fission cross section σ_f . In this case fission can also be induced by secondary neutrons and alpha particles. This effect would be enhanced by reactions in the foils used to degrade the carbon-ion energy. Thus corrections might be necessary in using this method. Pre-

liminary results from radiochemical measurements of σ_f give,²⁷ within the experimental errors, agreement with $\sigma_c(E_C)$ obtained in this work.

It is now possible to calculate the cross sections for the (C, xn) reactions of other heavy isotopes. In this case we assume, as a first approximation, that for isotopes of not too widely different atomic number the cross sections for compound-nucleus formation [$\sigma_c(E_C)$] are equal at the same value of the parameter $x = E_C / V_C$, (E_C, V_C previously defined). Accordingly we have calculated the cross section for the reaction $\text{Pu}^{242}(C, 4n)\text{Fm}^{250}$, using a nuclear temperature of 0.90 Mev, and the agreement with the experimental results was good as shown in Fig. 2. A recoil method similar to that for helium-induced reactions was used in these experiments.

It is reasonable to assume that similar treatment can be applied to predict cross sections of reactions, involving neutron evaporation, of other heavy ions with heavy isotopes.

B. $(C, \alpha 4n)$ Reactions

The cross-section curve for the $(C, \alpha 4n)$ reaction (shown in Fig. 1) seems to contain at least two components. There is a broad peak upon which a sharper one with a maximum at 76 Mev is superimposed. Energy considerations show that in both cases alpha particles rather than other charged particles must actually be emitted in the reactions at lower energies.

From our data it is not possible to arrive at definite conclusions concerning the mechanisms involved in the $(C, \alpha 4n)$ reaction. However, the following discussion represents our point of view on this question. The broad peak is probably a consequence of reactions that do not involve the evaporation of alpha particles from a compound system. An evaporation process should give a sharper peak. Furthermore, it should result in a sharp drop in the cross section below the threshold corresponding to a kinetic energy of the emitted alpha particle equal to the Coulomb barrier. For the $(C, \alpha 6n)$ reaction this threshold is 83 Mev (for the C^{12} ions), whereas we observe products from the reaction far below this energy. The drop in the $(C, \alpha 4n)$ curve below the evaporation threshold at 69 Mev might be due to the influence of the barrier on the C^{12} ions.

One possible explanation of the broad part of the $(C, \alpha 4n)$ reaction curve is obtained if we assume that alpha-particle structure exists in the carbon ion for sufficiently long periods of time to allow stripping or electric disintegration to occur. Disintegration of C^{12} into He^4 and Be^8 is actually observed in photographic emulsions.²⁸ In stripping, certain orientations would favor the penetration of the Be^8 , whereas the alpha particle would be prevented from amalgamation by the centrifugal and Coulombic barrier. In electric dis-

²⁷ S. G. Thompson (unpublished).

²⁸ James F. Miller, University of California Radiation Laboratory Report UCRL-1902, July, 1952 (unpublished).

²⁶ Vandenbosch, Thomas, Vandenbosch, Glass, and Seaborg, *Phys. Rev.* **III**, 1358 (1953).

integration in the Coulomb field, Be⁸ could enter the target nucleus whereas the alpha particle would be scattered. The alpha particle might carry off a wide range of energies, resulting in a broad peak for the (C, α n) cross-section curve. If now $\sigma(C,\alpha)$ is the total cross section for reactions involving (C, α) stripping, the cross section for the (C, α 4 n) reaction is

$$\sigma_{(C,\alpha 4n)} = \sigma(C,\alpha) \cdot (\bar{G})^4 P_{4n}. \quad (3)$$

$(\bar{G})^4$ is taken from data on the reaction Pu²⁴²(α ,4 n)Cm²⁴² to be 8×10^{-3} .¹² P_{4n} is always less than one and $\sigma_{(C,\alpha 4n)}$ is a measured value. At 76 Mev we obtain $\sigma(C,\alpha) > 10$ millibarns.

The sharp-peak component of the (C, α 4 n) curve might actually be due to the evaporation of alpha particles from the compound system. In this case, an order-of-magnitude estimate of the partial level width for alpha emission [$G_\alpha = \Gamma_\alpha/\Gamma_\alpha$] can be obtained. If the alpha particle is evaporated first, the cross section for the (C, α 4 n) reaction may be expressed as follows:

$$\sigma(C,\alpha 4n) = \sigma_c(E_C) G_\alpha (\bar{G})^4 \cdot P_{4n}. \quad (4)$$

Here, $(\bar{G})^4$ is again taken from data on the reaction Pu²⁴²(α ,4 n)Cm²⁴².¹² P_{4n} is always less than 1 and $\sigma(C,\alpha 4n)$ and $\sigma_c(E_C)$ are known values. At 76 Mev we obtain $G_\alpha > 0.01$. If the alpha particle is evaporated in a later step, \bar{G} for the neutron-level width becomes smaller and thus G_α becomes larger. If the alpha par-

ticle is evaporated after the neutrons, we obtain $G_\alpha > 0.04$.

The sharp-peak component could also be explained if in the electric disintegration the alpha particles carry off one-third of the kinetic, internal, and potential energy of the carbon ions.†

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† *Note added in proof.*—Preliminary measurements indicate that the maximum energy of the C¹² ions was 125 Mev rather than 120 Mev [John R. Walton, University of California (private communication, 1958)]. This will make a better agreement with helium induced reactions. In some recent experiments Flerov *et al.* [Academy of Atomic Energy, Moscow, USSR (private communication, 1958)] have measured excitation functions for C¹² induced reactions in U²³⁸ and found the peak for the (C,4 n) reaction to be at 69 Mev. The cross section for the reaction was twice as high as found in our experiments. The discrepancy can be ascribed to uncertainties in the beam intensities.

Energy Levels in F¹⁸ from the N¹⁴(α , α)N¹⁴ and N¹⁴(α , p)O¹⁷ Reactions*

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Elastically scattered alpha particles from N¹⁴ and ground-state protons from the N¹⁴(α , p)O¹⁷ reaction show resonances at 2.88-, 3.09-, 3.60-, 3.67-, 3.72-, 4.00-, 4.05-, 4.11-, 4.28-, 4.50-, and 4.55-Mev bombarding energies, corresponding to excited states of the F¹⁸ nucleus at 6.65, 6.82, 7.21, 7.27, 7.30, 7.52, 7.56, 7.61, 7.74, 7.91, and 7.95 Mev. Scattering-matrix analysis of the elastically scattered alpha particles, together with angular distributions of the reaction protons, indicate that the 6.65-, 6.82-, 7.21-, 7.27-, 7.30-, 7.52-, 7.91-, and 7.95-Mev states in F¹⁸ probably have angular momenta and parities of 1⁻, 2⁻, 4⁺, 1⁺, 3⁻, 3⁻, 2⁻, and 1⁺, respectively.

INTRODUCTION

OBSERVATIONS on scattered α particles and reaction protons from accelerated helium ions incident on nitrogen gas have been extended to 4.7 Mev. A number of narrow resonances have been observed in the energy range from 3.5 to 4.7 Mev. Phase-shift analysis of the elastic scattering was not attempted because of the complexity associated with spin 1.¹ Instead, an

attempt has been made to determine angular momenta and parities of the corresponding excited states of F¹⁸ from excitation curves at several angles in the immediate neighborhood of the resonances, using the scattering-matrix analysis as described by Blatt and Biedenharn.²

The reaction N¹⁴(α , p)O¹⁷ was the first α -induced nuclear transmutation observed by Rutherford.³ This reaction figured prominently in the early investigations

* Supported in part by the U. S. Atomic Energy Commission.

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¹ J. M. Blatt and L. C. Biedenharn, Phys. Rev. **86**, 399 (1952).

² J. M. Blatt and L. C. Biedenharn, Revs. Modern Phys. **24**, 258 (1952).

³ E. Rutherford, Phil. Mag. **37**, 581 (1919).