Ejection of Large Fragments in High-Energy Nuclear Reactions*

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The formation cross section for Na^{24} and Mg^{28} was measured in targets of Cu, Ag, Au, and U bombarded with protons and helium ions over the energy range 320-880 MeV. In addition, the kinetic energy of these products was measured by a thick-target, recoil-range technique at incident proton energies of 0.7 and 3.0 GeV and an incident helium-ion energy of 0.88 GeV. In both sets of properties there was found a close similarity in the results from helium-ion and proton bombardments. In the case of the proton bombardments the results were compared to the predictions of the conventional model of high-energy regions, i.e., a fast nucleonic cascade followed by a slower evaporation cascade. It was found that the conventional model cannot reproduce the results, which suggests that another reaction mechanism, fragmentation, is required. Possible views of a fragmentation mechanism are presented, and the implications of the experimental results for the nature of the fragmentation mechanism are explored,

I. INTRODUCTION

THIS paper describes a radiochemical study of the
production characteristics of the radionuclides
Na²⁴ and Mg²⁸ in targets of copper, silver, gold, and HIS paper describes a radiochemical study of the production characteristics of the radionuclides uranium bombarded with 700-MeV protons and with helium ions in the energy range of 320-880 MeV. The formation cross sections were measured by standard techniques. In addition, certain recoil properties of these products were measured by a thick-target technique. These recoil experiments provided a measure of the intrinsic kinetic energy of the products and of the center-of-mass velocity of the heavy progenitor of the Na²⁴ and Mg²⁸. The experimental quantities were compared with the predictions of various reaction mechanisms. The purpose of the investigation was to provide information on the mechanism of fragmentation, a fast violent process, in which a complex nucleus is divided into two or more complex aggregates of nucleons. Since the results have meaning chiefly when discussed in the context of previous studies of the interaction of highenergy particles with complex nuclei, we begin by reviewing certain features and conclusions of previous studies.

A commonly used description of high-energy reactions is a two-stage mechanism consisting of a fast cascade step followed by a slow evaporation step. In the first stage the incoming particle undergoes an elastic or inelastic collision with an individual nucleon in the nucleus. This collision may initiate a complex cascade during which a few high-energy nucleons escape from the nucleus and large amounts of energy are absorbed by the target nucleus. It is also possible that the bombarding particle escapes after only a single collision carrying off most of its original energy. In the general case a wide distribution of excitation energy is left in the collection of struck nuclei. In the second step the excitation energy of the struck nuclei is dissipated by successive evaporation of several neutrons, protons, and

* This report is a revision of the material published in the Ph.D. thesis of one of us, V. P. Crespo, previously issued as Lawrence Radiation Laboratory Report UCRL-9683, 1961. alpha particles. With lesser probability there may be evaporation of heavier aggregates of nucleons. In the case of the heavier elements de-excitation may also occur by nuclear fission.

This two-step model predicts only four categories of reaction products: (1) Prompt-cascade neutrons and protons emitted with a broad spectrum of kinetic energies ranging up to full energy of the bombarding particle. It is possible that alpha particles or more complex aggregates of nucleons existing as a transient substructure in the nucleus may participate in the highenergy cascade and be ejected from the nucleus. (2) Evaporated nucleons or nucleon aggregates emitted symmetrically in the center-of-mass system with an energy distribution characterized by a Coulombic barrier and a nuclear temperature. Neutrons, protons, and helium ions are most prominent but some contribution may be expected from fragments of higher *Z.* (3) Fission products. (4) Evaporation residues. These last are the stable or radioactive nuclides remaining at the end of the evaporation chain.

The detailed characteristics of the high-energy cascade have been calculated by Monte Carlo techniques for a representative group of target elements and bombarding particle energies.¹⁻⁵ The most extensive calculations are those of Metropolis et al.⁵ which covered two energy ranges—(1) bombarding energies for which only elastic collisions were significant and (2) bombarding energies up to 2 GeV in which inelastic, meson-producing, collisions are expected to occur in considerable frequency. Such calculations lead to predictions of cascade-particle frequency, angular distribution, and energy distribution which can be compared with data obtained in nuclear-emulsion studies of proton and alpha-particle tracks. When the energy of the bombarding particle is insufficient for significant meson production the agreement of the emulsion data with the

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² J. Combe, Nuovo Cimento 3, 182 (1956).

³ J. W. Meadows, Phys. Rev. 98, 744 (1955).

⁴ G. Rudstam, thesis, Uppsala, 1956 (uppublished).

⁴ S. Rudstam, thesis, Up

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calculations is quite close. In the higher energy ranges the agreement is only fair owing probably to errors in the input data on inelastic collisions for the Monte Carlo calculations.

The calculations of the cascade step also lead to a prediction of a set of excited nuclei and a distribution in excitation energy for those nuclei. These predictions cannot be used for a direct comparison with experimental data but they may be used as the input data for a calculation of evaporative de-excitation. Such calculations^{4,6-8} lead to a prediction of frequency and energy spectra of light evaporated particles, chiefly neutrons, protons, and alpha particles, which can be compared with the black prong data of nuclear emulsion studies. The calculations also lead to a prediction of the yields of heavy residual nuclei which can be compared to experimental cross-section data obtained by radiochemistry and mass spectrometry.

Miller and Hudis⁹ have thoroughly reviewed the fastcascade-plus-evaporative-de-excitation mechanism of high-energy reactions, published Monte-Carlo calculations of the two stages of the reaction, and the confrontation of the predictions with results obtained in emulsion and radiochemical experiments. In general, it has been concluded that there is good-to-excellent agreement of many of the experimental results with the predictions of the model for bombarding energies up to several hundreds of MeV. In the GeV energy range there is only fair agreement although much better agreement might be expected if better input parameters for inelastic collisions are found and used in a revised calculation.

We are not concerned here with a detailed examination of those features of high-energy reactions which seem explained or capable of explanation by the conventional description of the reaction mechanism. We wish instead to call attention to a growing body of phenomena which cannot be described adequately by the model and which suggest the necessity to extend its scope or to consider the existence of an additional reaction mechanism fundamentally different in nature.

The term fragmentation has been applied to this different mechanism or mechanisms. Fragmentation is some violent fast disruption of the nucleus which leads to the production of complex aggregates of nucleons in appreciable yield. Fragmentation may be a fission-like process in that two or more chunks of the nucleus may separate and achieve considerable kinetic energy by mutual Coulombic repulsion, but it differs from fission in speed, and in the angular, energy, and mass distributions of the products.

A. Emulsion Evidence for Fragmentation

Much of the information suggesting a fragmentation process has come from nuclear-emulsion studies in which light fragments with charge greater than two have been identified. This information is collected in an excellent review article by Perfilov, Lozhkin, and Shamov.¹⁰ A number of the more significant articles are cited below.11-23

The majority of the emulsion data pertain to the production characteristics of Li⁸ because of the easily identified "hammer tracks" resulting from the decay of Li⁸ into Be⁸ which instantly disintegrates into two alpha particles. However, there are some data, particularly in the Russian literature,¹⁰⁻¹⁴ in which tracks of fragments with *Z>3* have been studied.

The frequency of occurrence of these light fragments is a steep function of bombarding energy in the range 100 MeV to several GeV and the stars associated with fragment production contain a substantially larger number of cascade proton tracks than do the average of all stars. These facts indicate that fragment production is favored by very high energy transfers in the cascade step. The energy spectrum of these light fragments has the general appearance of an evaporation spectrum but it is in most cases too broad to be explained by any reasonable choice of evaporation parameters.15-19 In addition, several authors have reported a group of highenergy fragments many tens of MeV beyond the maximum of the evaporation spectrum.^{7,18,20} In some cases energies greater than 100 MeV have been found.^{13,19} These higher energy fragments have special characteristics. For example, Nakagawa, Tamai, and Nomoto¹⁸ reported that the yields of the highest energy group of Li⁸ fragments produced in the bombardment of emulsion with 6-GeV protons had a strong dependence on the number of cascade charged particles (grey tracks) but no dependence on the number of evaporated charged particles (black prongs) emitted from the same star.

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20 E, W, Baker and S, Katcoff, Phys. Rev, **123,** 641 (1961),

⁶ 1 . Dostrovsky, P. Rabinowitz, and R. Bivins, Phys. Rev. **Ill,** 1659 (1958); I. Dostrovsky, Z. Fraenkel, and P. Rabinowitz, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy* (United Nations, New York, 1958), Vol. 15, p. 301. 7 G. Friedlander, Z. Fraenkel, and I. Dostrovsky, Phys. Rev.

^{116,} 683 (1959); J. Hudis and J. M. Miller, *ibid.* **112,** 1322 (1958).

⁸ 1 . Dostrovsky, Z. Fraenkel, and J. Hudis, Phys. Rev. **123,** 1452 (1961); I. Dostrovsky, Z. Fraenkel, and P. Rabinowitz, *ibid.* **118,** 791 (1960).

⁹ J. M. Miller and J. Hudis, Ann. Rev. Nucl, Sci. 9, 159 (1959).

¹⁰ N. A. Perfilov, O. V. Lozhkin, and V. P. Shamov, Usp. Fiz. Nauk 60, 3 (1960) [translation: Soviet Phys.—Usp. 3, 1 (I960)]. 11 O. V. Lozhkin and N. A. Perfilov, Zh. Eksperim. i Teor. Fiz.

^{31,} 913 (1956) [translation: Soviet Phys.—JETP 4, 790 (1957)]. 12 V. S. Ostroumov and Iu.P. Iakovlev, Zh. Eksperim. i Teor.

Fiz. 35, 1358 (1958) [translation: Soviet Phys.—JETP 8, 949 (1959)]

¹³ O. Skjeggestad and S. O. Sorensen, Phys. Rev. **113,** 1115 $(1959).$

¹⁴ P. A. Gorichev, V. F. Darovskikh, O. V. Lozhkin, A. I. Obukhov, N. A. Perfilov, and U. P. Jakovlev, Phys. Rev. **126**, 2196
(1962) and P. A. Gorichev, O. V. Lozhkin, N. A. Perfilov, and
Yu. P. Yakovlev, Zh. Eksperim. i Teor. Fiz. 41, 327 (1961)
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^{780 (1958).} 19 S. O. Sorensen, Phil. Mag. 40, 947 (1949); 42, 188 (1951).

This suggested strongly that these Li⁸ fragments were not evaporated but were produced somehow in the cascade process. A similar result was reported by Goldsack, Lock, and Munir.²¹ These authors also pointed out that a lower energy group of Li⁸ fragments occurring in complex stars with more than 12 black prongs had an isotropic distribution with respect to the bombarding protons, whereas a higher energy group associated with less complex stars was peaked strongly forward.

Katcoff¹⁶ placed foils of Cu, Ag, Au, and U in a beam of 2.2-GeV protons and examined the properties of Li⁸ fragments ejected from the targets and caught in nuclear emulsions. The energy spectrum in the case of the silver target was approximately correct for an evaporation spectrum but in the other three cases it was too broad and had too much intensity in the higher energy regions.

In the examination of stars in nuclear emulsions it is sometimes observed that two or more fragments with $Z>2$ are emitted in a single nuclear disintegration.^{10,11,17} From evaporation theory such events should have an exceedingly low probability, but experimentally it is found that in several percent of the events containing any fragments whatsoever there is double or triple fragment emission.¹⁰

The evaporation model would predict a moderate favoring of emission in the forward direction with respect to the beam because of a small center-of-mass motion of the struck nucleus. Many authors found a more pronounced favoring of the forward hemisphere than seemed reasonable^{11,15} although Skjeggestad and Sörensen¹³ concluded that the forward peaking of Li⁸ fragments could be accounted for satisfactorily. Some like Baker and Katcoff²⁰ (emulsion plus 1-, 2-, and 3-GeV protons) and Perkins¹⁷ (emulsions plus cosmic rays) have noted a preference for sideways emission with respect to the beam in addition to the expected forward peaking.

Some authors have reported favored directions of emission of fragments with respect to the tracks of alpha particles, protons, residual heavy nuclei, and other fragments. This suggests that the fragment and the entity making the correlated track are emitted simultaneously.

Several authors have considered and rejected^{11,15,18,19} the hypothesis that Li⁸ and other light fragments are produced by a direct elastic collision of the incoming particle with a Li⁸ or similar subgroup in the nucleus. Such a model is not compatible with the observed lack of correlation of the energy of the fragment and the angle of emission. Nor can it account for multiple fragment emission and other features.

The over-all conclusion from these emulsion studies

is the following. Although the conventional reaction mechanism of cascade plus evaporation undoubtedly explains the formation of part, perhaps in many cases the major part, of the light fragments, there is strong evidence for the existence of an additional process for their formation.

B. Radiochemical Evidence for Fragmentation

Let us now consider the radiochemical evidence which consists chiefly of the formation cross section and excitation functions for several radioactive nuclides in the mass range 7 to \sim 40 produced in various target elements bombarded with high-energy particles. Some of the nuclides studied—such as He⁶, Be⁷, C¹¹, and N¹³ overlap the range of mass numbers covered in the emulsion studies. Others—such as, F¹⁸, Na²², Na²⁴, Mg²⁸, Si³¹, P³², and P³³—are considerably heavier in mass. The excitation functions for such products show a sharp rise starting at several hundred MeV and continuing to about $2 \text{ GeV}, ^{24-26}$ above which there is a sharp leveling off in cross section.

Several authors^{26,27} have noted an interesting variation in the cross sections of such products as F^{18} , Na²⁴, and P³² as a function of the target mass number. Later on we shall discuss our own results bearing on this point in connection with Figs. 5 and 6. Up to a target mass number of \approx 150 there is a decrease in cross section, whereas above mass 180 there is a marked increase. It seems difficult to explain the yield increase for the higher mass targets by a spallation mechanism, and Caretto, Hudis, and Friedlander²⁶ attributed it to fragmentation.

The curves displaying cross section versus mass number for products formed in heavy target elements bombarded with GeV protons also have a peculiar form which has been cited as evidence for a fragmentation mechanism. Wolfgang *et at.* and Friedlander and Yaffe^{25,28} studied the interaction of lead with protons of 1.0- to 3.0-GeV energy. A rather remarkable feature of the total isobaric cross sections is their near constancy over the entire range of masses. From mass 20 up to the mass of the target the yields differ only by a factor of 10 or 20. This curve is entirely different from those determined for bismuth targets bombarded with 340- or 480-MeV protons. In the latter cases there is a prominent group of fission products and a clearly identifiable group of spallation products (evaporation residues) close in mass to the target mass. The yields of products in the latter group drop off rapidly as the product mass number decreases.

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²² E. Pickup and L. Voyvodic, Can. J. Res. 28A, 616 (1950). 23 F. S. Rowland and R. L. Wolfgang, Phys. Rev. **110,** 175

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²⁴ E. Baker, G. Friedlander, and J. Hudis, Phys. Rev. **112,** 1319 (1958).

²⁵ R. Wolfgang, E. W. Baker, A. A. Caretto, J. B. Cumming, G. Friedlander, and J. Hudis, Phys. Rev. **103,** 394 (1956). 26 A. A. Caretto, J. Hudis, and G. Friedlander, Phys. Rev. **110,**

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²⁷ A. K. Lavrukhina, L. P. Moskaleva, L. D. Krasavina, and I. M. Grechishcheva, Soviet J. At. Energy 3, 1087 (1957). 28 G. Friedlander and L. Yaffe, Phys. Rev. **117,** 578 (1960).

FIG. 1. The cross section assembly. T_1 , T_2 , T_3 , and T_4 are the target foils separated by 0.003-in.-thick Mylar spacers (My); Al_{mon} is the Al monitor foil. The other foils are guard foils.

A consideration of the radiochemical data in the leadbismuth targets led Wolfgang *et al?^h* to postulate a fast fragmentation of the nucleus resulting from local heating caused by the production and reabsorption of pions.

C. Purpose and Scope of Present Work

The present work was undertaken in the belief that additional data on the formation properties of Na²⁴ and Mg²⁸ would confirm the need for the postulated fragmentation process and throw some light on its nature. Na²⁴ and Mg²⁸ were chosen because of their convenient radiochemical properties and because there seemed little reason to believe that they could be formed in high yield either as evaporated particles or cascade-evaporation residues in targets as heavy as silver, gold, or uranium. In the case of copper targets it seemed likely that they would be produced chiefly as evaporation residues, but it was deemed desirable to verify this. The existence of a considerable amount of Na^{24} cross-section data for proton-induced reactions was also an advantage in the discussion of our results. Except for these features, there is nothing special about Na^{24} and Mg^{28} ; hence, they may serve as representatives of the behavior of a whole class of products of roughly similar mass produced in highenergy reactions.

In separate experiments, the target elements were bombarded with 700-MeV protons, and with 320-, 500-, 700-, and 880-MeV helium ions and the formation cross sections were measured by the radiochemical techniques given in detail below. The comparison of the helium-ioninduced production curves with those for protoninduced production was thought to be particularly significant in evaluating the contribution of meson effects to fragmentation yields. It was anticipated that helium ions of a given energy would be much less effective in creating mesons in the targets nuclei than protons of the same energy. Hence, if fragments such as Na²⁴ are, indeed, produced exclusively by a mechanism involving production and reabsorption of mesons then the production cross section should be substantially less when helium ions are substituted for protons.

In addition to cross-section measurements, our experiments also included a study of the recoil properties of the fragments.

Our plan in the analysis of the cross section and recoil range results was to assume at first the correctness of the conventional two-stage description of high-energy reactions. We then made a number of qualitative and quantitative comparisons of the predictions of this model with those characteristics which could be extracted from our experimental data under the assumption that the model was correct. A major fraction of this paper consists of the development of this comparison. It will be found that there appear to be internal contradictions of fundamental importance when the data are treated in the framework of the conventional model particularly at the higher bombarding energies. We will conclude that the conventional view of a fast stage plus slow stage cannot account for the production of such nuclides as Na²⁴ and Mg²⁸ in targets of silver, gold, and uranium and may not account for it even in targets of copper. We then consider other possible methods for the production of such fragments.

II. CROSS-SECTION DATA

A. **Experimental**

Target Assemblies

The target assembly used in the determination of cross sections consisted of a stack of $1.5\text{-cm}\times2\text{-cm}$ foils arranged as shown in Fig. 1. The target foils designated T_1 , T_2 , T_3 , and T_4 were separated from one another by 0.003-in. Mylar foils (designated My). All cross sections were measured relative to Na²⁴ produced in the aluminum monitor foil (Al-Mon). The number of target foils used in any particular experiment varied from one to four. The copper target foils were 0.002 in. thick; all others were 0.001 in. thick. Spectrochemical analysis of the target foils showed the following levels of contamination: copper targets-0.03% Al; silver targets-0.1% Cu and 0.02% Al; gold targets-0.3% Cu; uranium targets-100 ppm of Si and 10 ppm of Cu, Co, and Mg; aluminum targets- 0.05% of Cu and Fe. It was of critical importance to align exactly the different foils in the stack because most of the beam struck the leading edge. To insure proper alignment the leading edge was machined after the stack was fastened to the target holder. The stack was then wrapped in 0.001-in.-thick Mylar film.

All foils were cleaned before mounting for irradiation. Copper and gold foils were cleaned with dilute nitric acid; uranium foils with 2 to *6M* nitric acid and silver foils with ammonia. All foils were then washed with acetone and rinsed with distilled water.

Irradiations and Beam Monitoring

The irradiations were performed at the 184-in. cyclotron and the bevatron. Bombardment times varied from 1 to 1.5 h.

The Na²⁴ produced in the aluminum monitor foils by the reaction $A^{27}(p,3pn)$ or $A^{27}(\alpha,\alpha^2pn)$ was determined by cutting up the monitor foils and mounting them in a fashion to reproduce as closely as possible the conditions used for activity measurements in the samples isolated

Target	Product	320-MeV α	Protons 700-MeV p 5.7-GeV p				
Cu	Na^{24} Mg^{28} Mg^{28}/Na^{24} Na ²⁴ /Na ²²	$8.26 \times 10^{-2}(1)$ $1.15 \times 10^{-2}(1)$ 0.14	$(2.80 \pm 0.13) \times 10^{-1}(3)$ $0.37 \times 10^{-1}(1)$ 0.13	$(6.98 \pm 0.13) \times 10^{-1}(3)$ $0.91 \times 10^{-1}(1)$ 0.13	$1.38 + 0.57(3)$ 0.19(2) 0.13	$3.68 \times 10^{-1}(1)$ $0.49 \times 10^{-1}(1)$ 0.12 7.44 ^b	1.65c
Аg	Na ²⁴ Mg^{28} Mg^{28}/Na^{24}	$3.25 \times 10^{-2}(1)$ $0.39 \times 10^{-2}(1)$ 0.12	$(9.52 \pm 0.72) \times 10^{-2}(3)$ $1.10\times10^{-2}(1)$ 0.12	$(2.27 \pm 0.005) \times 10^{-1}(3)$ $0.26 \times 10^{-1}(1)$ 0.12	$(4.46\pm0.02)\times10^{-1}$ (3) $0.49 \times 10^{-1}(2)$ 0.11	$1,00\times 10^{-1}(1)$ $0.12 \times 10^{-1}(1)$ 0.12	
Au	Na ²⁴ Mg^{28} Mg^{28}/Na^{24} Na ²⁴ /Na ²²	$3.23 \times 10^{-2}(1)$ 2.07×10^{-2} (1) 0.64	$(10.68 \pm 0.28) \times 10^{-2}(3)$ $6.24 \times 10^{-2}(1)$ 0.58	$(3.08 \pm 0.87) \times 10^{-1}(3)$ $1.02 \times 10^{-1}(1)$ 0.33	$(5.94 \pm 0.16) \times 10^{-1}(3)$ $1.98 \times 10^{-1}(2)$ 0.33	$1.35 \times 10^{-1}(1)$ $0.54 \times 10^{-1}(1)$ 0.23	5.29
U	Na ²⁴ $\rm Mg^{28}$ Mg^{28}/Na^{24} Na ²⁴ /Na ²²	$9.41 \times 10^{-2}(1)$ $6.42 \times 10^{-2}(1)$ 0.68	$(21.02 \pm 1.38) \times 10^{-2}(2)$ $12.29 \times 10^{-2}(1)$ 0.58	$(5.02 \pm 0.05) \times 10^{-1}(2)$ $2.38 \times 10^{-1}(1)$ 0.47	$(8.75 \pm 0.25) \times 10^{-1}(3)$ $4.50 \times 10^{-1}(2)$ 0.51	$2.30\times10^{-1}(1)$ $1.15 \times 10^{-1}(1)$ 0.32	5.004

TABLE I. Formation cross sections (in mb) of Na²⁴ and Mg²⁸ from various target elements.⁸

a Standard error is quoted if three replicate runs were made. The principal source of absolute error is uncertainty in the monitor cross sections as discussed in text. Reference 34.

© From D. W. Barr, Laurence Radiation Laboratory Report UCRL-3793, 1957 (unpublished).
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radiochemically from the target. The monitor cross sections were taken as 10.7 mb for $Al^{27}(p,3pn)Na^{24}$ and as 24.0 mb for $Al^{27}(\alpha,\alpha^2pn)Na^{24}$ for all energies. The proton-induced cross section has been determined at several bombardment energies²⁹ and is rather well established. The value of 24.0 mb for the alpha-induced reaction is that measured at the single energy of 380 MeV.30,31 The cross section is not expected to change drastically between 380 and 880 MeV, but an uncertain error is associated with the monitor cross section and with the cross sections reported below in targets bombarded with the higher-energy helium ions.

Treatment of Targets after Bombardment

After bombardment the metal foils were dissolved in an appropriate acid in the presence of a few milligrams of sodium and magnesium carrier. By means of the radiochemical procedure outlined in the Appendix, sodium and magnesium fractions were recovered from the solution in a high state of radiochemical purity. In the final step of the magnesium procedure an 8 hydroxyquinoline compound was precipitated, filtered, dried, and weighed to determine the percentage chemical recovery. Similarly, in the final step of the sodium procedure sodium chloride was precipitated from a solution of butanol saturated with hydrochloric acid.

The activity of the Na²⁴ and Mg²⁸ samples was measured in end-window proportional counters over a period of days. The 15-h decay of Na²⁴ could easily be resolved from the almost negligible background in the sodium fractions. The 21-h Mg²⁸ activity was clearly resolved from the decay of the magnesium samples, but in some experiments there was some contamination by longerlived activity. Activity-measurement techniques and corrections are discussed in the Appendix.

B. Results and Discussion

Cross-section results for the different targets and bombarding particles are listed in Table I. The ratio of the cross sections $\sigma_{Mg}^{28}/\sigma_{Na}^{24}$ is also tabulated. The number of determinations of any particular cross section is given in parentheses after the cross-section values. The numbers given are average values and the errors quoted are the standard errors.

The Na²⁴ and Mg²⁸ cross sections are plotted in Figs. 2 and 3 versus alpha-particle bombarding energy for the different targets. Figure 4 is a similar plot of Na²⁴ cross sections from proton bombardments; most of the points on this plot are taken from data in the litera-

FIG. 2. Excitation functions for the production of Na²⁴ from He-ion bombard ments of Cu, Ag, Au, and U.

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³⁰ M. Lindner and R. Osborne, Phys. Rev. 91, 342 (1953). 31 W. E. Crandall, G. P. Millburn, R. V. Pyle, and W. Birnbaum, Phys. Rev. 101, 329 (1956).

FIG. 3. Excitation functions for the production of Mg²⁸ from He-ion bombard ments of Cu, Ag, Au, and U.

	Yield Na 24 (700-MeV He)	Yield Mg^{28} (700-MeV He)
Target	Yield Na ²⁴ (700-MeV ϕ)	Yield Mg ²⁸ (700-MeV p)
Сu	1 Q	1.85
	2.27	2.17
Ag Au	2.28	1.85
T	2.18	2.07

TABLE II. Comparison of yields of Na²⁴ (and Mg²⁸) in targets bombarded with 700-MeV protons and helium ions.

which was observed earlier in the case of proton bombardments by Caretto, Hudis, and Friedlander²⁶ is seen to occur also for helium-induced reactions.

We call attention to the marked over-all similarity of the results for the proton and helium ion bombardments: namely, the thresholds, shapes of the excitation functions, and the yield variations with target mass number. These similarities suggest strongly that the helium ions and the protons generate a cascade in the nucleus of very similar character.

In the framework of the conventional two-stage reaction model the high threshold for the production of Na²⁴ and Mg²⁸ and the steady rise of the cross section up

FIG. 4. Excitation functions for the production of Na²⁴ from proton bombardments of Cu, Ag, Au, and U. Data from 1 to 5.9 GeV from Ref. 26. Data for Cu are from Refs. 27, 32-34, and for Ag from Refs. 35 and 36, for Au from Ref. 27 and for U from Refs. 37 and 38. The 700-MeV points are from this work.

ture.^{26,27,32-38} The principal feature we note on all three figures is that the probability of formation remains low for all targets until the bombarding energy reaches some hundreds of MeV. Then it increases rapidly but, for protons, levels off again in the GeV range of energies. Similar results have been observed for other fragments^{23-28,39} (Be⁷, F¹⁸, P³², P³³, and others).

At a given energy of the bombarding particle the absolute yields of Na²⁴ and Mg²⁸ are higher by a factor of about 2 when helium ions are substituted for protons as the bombarding particle. Table II summarizes numerical values for 700-MeV particles. This observation is subject to the large uncertainty in the monitorreaction cross section for 700-MeV helium ions, but it is not likely that this cross section is off by a factor of 2. The values of $\sigma_{Mg^{28}}/\sigma_{Na^{24}}$ are larger for the heavier targets $(Au-U)$ than for the lighter ones $(Cu-Ag)$ and are essentially independent of the bombarding energy.

The results are replotted in Figs. 5 and 6 to show the variation of cross section with the mass of the target. The turn-up of the yield at the higher target masses,

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39 N. A. Perfilov, N. S. Ivanova, O. V. Lozhkin, M. M. Makarov, I. V. Ostroumov, Z. I. Solov'eva, and V. P. Shamov, Zh. Eksperim. i Teor. Fiz. 38, 345 (1960) [translation: Soviet Phys.—JETP 11, 250 (I960)].

 $Na²⁴$ 50.62 LC fa for Na²⁴ _
700-MeV ი 700-MeV n section ,,
500-MeV o ١ö Cross **120-MAV** 100 150 200 250 Mass number of target

to the GeV range of energies means that only those prompt cascades which deposit greater than 250 MeV of energy are effective in producing Na^{24} and Mg^{28} . It means also that those cascades which deposit substantially more than 250 MeV are most effective in producing these nuclides.

Porile and Sugarman⁴⁰ have analyzed cross-section results in a more formal way by writing down the expression.

$$
\sigma_A(E_B) = \int_0^{E_{\text{max}}*} \sigma_g N(E^*, E_B) F_A(E^*) dE^*,
$$

where the symbols have the following meaning.

 $\sigma_A(E_B)$ is the cross section for the production of a product *A* when the energy of the bombarding particle is E_B , σ_g is the total-reaction cross section. $N(E^*, E_B)$ is the fraction of nuclei left with an excitation energy E^* at the end of the fast cascade when the bombarding energy is E_B . $F_A(E^*)$ is the fraction of the nuclei, excited to *E** at the end of the fast cascade, which deexcite in such a way as to form final product *A.* It is important to re-emphasize at this point that the model under discussion assumes that the excitation energy and the charge and mass number of the nucleus at the end of the prompt cascade completely determine the spectrum of final products. The *FA* function may be expected to be zero below some threshold, then rise to a maximum value, and, finally, perhaps turn over and decrease at higher values of E^* .

To get some notion of the shape of the $N(E^*,E_B)$ function we can examine the results of the Monte-Carlo cascade calculations of Metropolis *et al.^h* In Fig. 7 we show the distribution found by these authors for the case of cerium, bismuth, and uranium bombarded with high-energy protons. We note that there is a very broad energy distribution in the struck nuclei from zero energy up to the energy of the bombarding particles. The only part of this distribution which is effective in the production of such nuclides as Na²⁴ is the high-energy tail.

When the bombarding energy is raised, the curve in the region of high excitation rises and is extended to higher energies.

Because the energy distribution of the residual nuclei at the end of the prompt cascade is shifted only slowly with bombarding energy, and because the excitation function for the measured products rises rapidly with *EB* above a very high threshold, it follows that the $F_A(E^*)$ function is rapidly rising with E^* and that it is only those few cascades which lead to the highest values of E^* which are contributing the main part to the Na²⁴ and Mg²⁸ cross sections.

If one calculates or assumes an *N{E*,EB)* distribution and a total-reaction cross section one can work backward from the experimental value of the Na^{24} or Mg^{28} production excitation functions to determine a branching ratio function, $F_A(E^*)$. Once the values of N and F have been fixed it is possible to calculate the average excitation energy, \vec{E} , of struck nuclei, which ultimately disintegrate to produce a product of mass *A,* by the

FIG. 7. Gross distribution of excitation energies *N(E*)* calculated by Metropolis *et al.* (Ref. 5) for bombardments of cerium, bismuth, and uranium targets with high-energy protons.

⁴⁰ N. T. Porile and N. Sugarman, Phys. Rev. 107, 1422 (1957).

FIG. 8. The recoil assembly. The recoils generated in the target foil T were caught in the catcher foils a and a'. Foils b were used as blank foils; foils c were guard foils. All the catcher foils were 0.003-in.-thick Mylar.

following expression

$$
\vec{E} = \int_0^{E_{\text{max}}^*} \sigma_g E^* N(E^* , E_B) F_A(E^*) dE^* / \int_0^{E_{\text{max}}^*} \sigma_g N(E^* , E_B) F_A(E^*) dE^*.
$$

We have carried through such an analysis for our results and conclude that for any reasonable form of $N(E^*, E_B)$ which is nonvanishing up to the bombarding energy (i.e., a form similar to the high-energy portions of the curves in Fig. 7), the average excitation energy for the production of Na²⁴ in targets of copper, silver, gold, and uranium bombarded with 700 protons lies between 500 and 700 MeV in all cases. In the case of bombarding energies of 3 GeV the required average excitation energies are even higher.

This result in itself puts severe strain on the basic assumptions of the conventional reaction model as it is not at all clear that nuclei excited to such huge energies can exist long enough to undergo de-excitation in a way adequately described by a statistical model of evaporation of small particles or clusters. In the case of copper targets these excitation energies are equivalent to the total binding energy in the nucleus.

It is possible to deduce that the $F_A(E^*)$ curves for the four target elements are very similar to each other by the following line of reasoning. First of all, we note that the published $N(E^*, E_B)$ spectra of Metropolis *et al.*^{*b*} have nearly the same shape for all target elements *for the deposition energy region above 50 MeV* except for a proportionality constant. (See, for example, Fig. 15 in Ref. 5.) That is,

$$
N(E^*,E_B)_{\text{Target 1}} = kN(E^*,E_B)_{\text{Target 2}}.
$$

Secondly, we note from Fig. 4 that the excitation functions for the production of Na²⁴ in targets of silver, gold, and uranium bombarded with proton are similar; the excitation function in the case of copper targets rises faster with bombarding energy up to 3 GeV and at higher energies stays more nearly constant than the corresponding excitation functions for silver, gold, and uranium. If the assumption is correct that the $N(E^*,E_B)$ distributions for large E^* differ only by a multiplicative constant then one expects the functions $F_A(E^*)$ for Na²⁴ production from silver, gold, and uranium also to differ

only by a constant factor and the function $F_A(E^*)$ for Na²⁴ production in copper targets to be somewhat displaced toward lower energies in relation to the $F_A(E^*)$ functions for the other targets. We shall see below that these conclusions are inconsistent with those derived from the recoil range results if we try to explain all results by the assumed two-stage mechanism.

III. RECOIL MEASUREMENTS

A. Basic Assumptions

Because of limitations imposed by beam intensities, low cross-section values, and permissible target thicknesses it proved most convenient to use a very simple, integral-range technique originated by Sugarman *et al.Al* for our measurements of recoil properties. In this method a thick target of precisely measured thickness is sandwiched between two Mylar catcher foils whose thickness is greater than the range of the products being measured. The recoil target assembly is shown in Fig. 8. The target foil (T) had dimensions 1.5 cm \times 2 cm while the other foils had dimensions $1.9 \text{ cm} \times 2.2 \text{ cm}$. The Mylar foils (My) were 0.003 in. thick (much thicker than the recoil range of Na^{24} or Mg^{28}). Blank foils were included to serve as a measure of the production of Na²⁴ and Mg^{28} by activation of impurities in the Mylar.

After the irradiation of this foil stack a radiochemical analysis was made to determine the amount of Na²⁴ and Mg²⁸ present in the target and in the upstream and downstream catcher foils. The radiochemical procedures, the methods of determination of radioactivity in the samples, and the corrections to the data are discussed in the Appendix.

From the corrected decay-rate data it was possible to compute the fractions of the total Na²⁴ or Mg^{28} activity which remains in the target foil, and which recoils into the upstream and downstream catchers. These fractions are designated F_T , F_B , and F_F , respectively. Some foil stacks were also exposed at 10 deg to the beam, and in these experiments the fractions of activity left in the target and recoiling in a perpendicular direction into the two side foils were determined. These fractions are designated F_{TP} , F_{PB} , and F_{PF} , respectively. The *F* (for forward) and *B* (for backward) in the latter subscripts allow for the fact that the bombardment stack is not precisely parallel to the beam. If the foil stack were precisely parallel F_{PF} and F_{PB} would be equal. The perpendicular recoil fraction was taken as *F^P* $=\frac{1}{2}(F_{PF}+F_{PB})$.

The analysis of the recoil results in terms of the conventional two-step mechanism depends upon the following very general assumptions, (a) The bombarding particle imparts to the target nucleus a velocity, v, which has a laboratory system component, *vn,* along the beam direction and a component, *Vi,* in a direction perpendicular to the beam, (b) When this struck nucleus

⁴¹ N. Sugarman, M. Campos, and K. Wielgoz, Phys. Rev. **101,** 388 (1956); N. T. Porile and N. Sugarman, *ibid.* **107,** 1410 (1957).

$\mathbf{1}$	2	3	4	5	6	7	8	9	10 \boldsymbol{v}	11 \bar{P}_T	12
Tar- get	No.	Beam	$W F_F$ (mg/cm ²)	$W F_B$ (mg/cm ²)	F_F/F_B	η	R_0 (mg/cm^2) (MeV)	E	(MeV/	(amu amu) ^{1/2} MeV) ^{1/2} \bar{P}_T/P_{CN}	
Cu	2	880-MeV α 700-MeV p $3-GeV$ p 320-MeV α	$1.678 + 0.055$ 1.184 1.036 (2, 440)	0.202 ± 0.000 0.352 0.370 (0.338)	8.23 ± 0.29 3.37 2.79 (7.19)	0.42 0.25 0.22	2.99 2.81 2.63	14.4 13.2 12.1	0.46 0.27 0.22	29.4 16.9 13.8	0.33 0.38 0.10
Ag		880-MeV α 700-MeV ρ 3 -GeV p	$3.345 + 0.072$ 2.253 1.848	0.645 ± 0.002 0.969 0.813	5.09 ± 0.13 2.32 2.27	0.33 0.18 0.17	6.88 6.18 5.10	30.2 25.7 20.4	0.53 0.26 0.23	57.3 28.1 24.4	0.64 0.63 0.21
Au		880-MeV α 700-MeV ρ 3-GeV p 6.2, 4.5-GeV p 320-MeV α	6.576 ± 0.096 4.728 4.560 4.452 (5.370)	$2.166 + 0.072$ 2.796 2.184 2.202 (2.718)	3.03 ± 0.06 1.68 2.09 2.01 (2.01)	0.23 0.11 0.16	16.22 14.92 13.05	60.0 54.0 45.6	0.52 0.23 0.30	102.6 46.4 59.9	1.15 1.05 0.52
\mathbf{U}	5	880-MeV α 700-MeV ρ 3 -GeV p	6.200 ± 0.035 4.165 ± 0.081 5.450	$2.230 + 0.024$ 2.520 ± 0.043 2.635	$2.77 + 0.03$ 1.64 ± 0.006 2.06	0.21 0.106 0.15	15.82 13.30 15.66	58.1 46.1 56.6	0.47 0.21 0.33	112.6 49.3 79.5	1.26 1.12 0.68

TABLE III. Summary of Na²⁴ recoil results from the forward-backward experiments.⁸

* Errors quoted in columns 4-6 are the standard deviations for replicate runs. Over-all errors are discussed in the Appendix.

disintegrates to form Na^{24} or Mg^{28} the breakup process provides an additional velocity component \bar{V} . This velocity V reflects the intrinsic kinetic energy of the fragment in the frame of reference of the moving nucleus. (c) The range is given by $\mathbf{R} = k(|\mathbf{V}+\mathbf{v}|)^N$, where k and *N* are constants to be evaluated empirically from rangeenergy data on fragments of similar size, charge and energy. (d) The angular distribution $W(\theta)$ of V in the moving frame is given by

$$
W(\theta) = a + b \cos^2 \theta.
$$

(e) The magnitudes of **v** and **V** are unique and (f) the path of the fragment is a straight line. [A more precise statement of assumption (f) is that the scattering of the observed fragments is similar to that of the energetic fragments of known energy which were used to calibrate the range-energy curve.]

Sugarman⁴² and Winsberg⁴³ have derived the necessary equations for the analysis of these experiments with the above assumptions. We use the notation $R_0 = kV^N$, $\eta_{11} \equiv v_{11}/V$, and $\eta_1 \equiv v_1/V$. If we neglect terms of order larger than η_{11}^2 , η_1^2 , and $(b/a)^1$ we obtain the following set of equations.⁴²

$$
F_F = \frac{R_0}{4W} \left\{ 1 + \frac{1}{6} \frac{b}{a} + \eta_{11} \left[\frac{2}{3} (N+2) + \frac{b}{a} \frac{(N-1)}{45} \right] + \eta_{11}^2 \left[\frac{(N+1)^2}{4} + \frac{b}{a} \frac{N^2 - N - 4}{12} \right] + \eta_{12}^2 \left[\frac{N^2 - 1}{8} + \frac{b}{a} \frac{N - 1}{12} \right] \right\}, \quad (1)
$$

$$
F_B = \frac{R_0}{4W} \left\{ 1 + \frac{1}{6} \frac{b}{a} - \eta_{11} \left[\frac{2}{3} (N+2) + \frac{b}{a} \frac{(N-1)}{45} \right] + \eta_{11}^2 \left[\frac{(N+1)^2}{4} + \frac{b}{a} \frac{N^2 - N - 4}{12} \right] + \eta_{12}^2 \left[\frac{N^2 - 1}{8} + \frac{b}{a} \frac{N - 1}{12} \right] \right\}, \quad (2)
$$
and

and

$$
F_P = \frac{R_{0P}}{4W} \left\{ 1 - \frac{1}{12} \frac{b}{a} + \eta_2 \frac{(N+1)}{16} \left[(3N+1) + \frac{b}{a} \left(\frac{7-3N}{6} \right) \right] + \eta_{11} \frac{(N-1)}{8} \left[(N+1) + \frac{b}{a} \left(\frac{N-5}{6} \right) \right] \right\}. \quad (3)
$$

Since there are four unknown quantities $(\eta_{11}, \eta_1, R_0, \eta_2)$ and b/a) and only three equations it is necessary to introduce at least one additional assumption. We chose to proceed by assuming that $\eta_1 = 0$ and $\bar{b}/a = 0$ and, hence, setting $\eta_{11} = \eta$ and $v_{11} = v$. These additional assumptions introduce only small errors in the calculated values of *V* and *Ro* and do not alter the conclusions. The effects of these assumptions are discussed in Appendix C.

In Appendix D we show that the value of $N=1.5$ fits best in the equation, $R_0 = kV^N$ for Na²⁴ and Mg²⁸ stopped in Cu, Ag, Au, and U. The proper values of the constant *k* are also given the Appendix D.

The working equations then reduce to

$$
WF_F = \frac{1}{4}R_0(1+2.333\eta+1.562\eta^2), \qquad (4)
$$

$$
W(F_F - F_B) = 1.166 \eta R_0, \qquad (5)
$$

$$
WF_P = \frac{1}{4} R_{0P} (1 + 0.156\eta^2). \tag{6}
$$

⁴² N. Sugarman (private communication); N. Sugarman and J.

Panontin (to be published). 43 L. Winsberg, University of California Lawrence Radiation Laboratory Report, UCRL 8618, 1959, pp. 44-52 (unpublished).

	2	3	4	5	6		
Target	Number of det.	Bombarding particle and its energy	WF_{PF} (mg/cm ²)	WF_{PB} (mg/cm ²)	R_{0P} (mg/cm ²)	R_0/R_{0P}	
Cu		880-MeV α	0.818	0.642	2.84	1.05	
		700-MeV p	0.664	0.630	2.56	1.10	
		3-GeV \bar{p}	0.742	0.576	2.62	1.00	
Ag		880-MeV α	1.866	1.494	6.60	1.04	
		700-MeV p	1.506	1.443	5.86	1.05	
		3-GeV \dot{p}	1.335	1.155	4.95	1.03	
Au		880-MeV α	4.218	3.498	15.30	1.06	
		700-MeV p	3.816	3.720	15.04	0.99	
		3-GeV p	3.702	3.198	13.73	0.95	
U	2	880-MeV α	4.025	3.285	14.51	1.09	
			± 0.001	± 0.169			
		700-MeV p	3.340	3.235	13.11	1.01	
		$3-GeV \, p$	4.140	3.515	15.24	1.03	

TABLE IV. Summary of Na²⁴ recoil results from the perpendicular experiments.⁸

* Over-all errors are discussed in the Appendix.

B. Recoil Results

Tables III and IV contain the Na²⁴ results and Tables V and VI the Mg^{28} results. Tables III and V contain the following information. Columns 1, 2, and 3 give the target, the number of determinations and the bombarding particles and their energy. Columns 4 and 5 list the products WF_F and WF_B needed for substitution in Eqs. (4) and (5). Columns 7 and 8 gives the quantities η and R_0 derived from Eqs. (4) and (5). The range values R_0 are converted into energy values $(E = \frac{1}{2}AV^2)$ by use of the range-energy expression $R_0 = kV^{1.5}$ whose justification is discussed in the Appendix. These energy values are listed in column 9.

The average velocity, v_{11} , imparted to the progenitor of the fragment is given by $v_{\text{H}} = \eta(2E/A)^{1/2}$. This velocity is quoted in the tables and in Figs. 10 and 11 in units of $(MeV/amu)^{1/2}$.

The measured and derived quantities from the perpendicular experiments are tabulated in Tables IV and VI.

C. Discussion of Recoil Results

We start our discussion of the recoil results with a consideration of the kinetic-energy values (E) of Na²⁴ and Mg²⁸ . A glance at the tables indicates that the kinetic energy of these products is quite high, comparable indeed to the kinetic energy observed for fission products of heavy elements. As a measure of comparison we may compute the Coulomb energy which such a product might obtain by a fission-type division of the

1	$\overline{2}$	3	$\overline{4}$	5	6	$\overline{7}$	8	9	10 $\boldsymbol{\eta}$	$\frac{11}{P_T}$	12
Target	No.	Beam	WF_F (mg/cm ²)	$W F_B$ (mg/cm ²)	F_F/F_B	η	R_0 (mg/cm ²)	E (MeV)	(MeV) $amu)^{1/2}$	(amu MeV ^{1/2}	\bar{P}_T/P_{CN}
Cu	$\overline{2}$	880-MeV α	1.546 ± 0.013	0.176 ± 0.011	8.80 ± 0.66	0.43	2.70	14.5	0.44	28.2	0.31
		700 MeV p	1.122	0.272	4.13	0.29	2.47	13.1	0.29	182	0.41
		3 -GeV \bar{b}	0.990	0.328	3.02	0.23	2.44	12.9	0.22	14.2	0.12
Ag	$\overline{2}$	880-MeV α	3.042 ± 0.050	0.669 ± 0.014	4.54 ± 0.03	0.31	6.50	32.7	0.48	51.6	0.58
		700 MeV p	1.995	0.897	2.22	0.17	5.53	26.6	0.23	25.3	0.57
		$3-GeV$ $\dot{\theta}$	1.749	0.801	2.18	0.16	4.92	22.9	0.21	22.8	0.19
Au	3	880-MeV α	6.744 ± 0.221	2.520 ± 0.041	2.67 ± 0.07	0.21	17.49	77.0	0.48	95.6	1.07
		700-MeV p	5.484	3.570	1.53	0.09	18.02	80.3	0.22	42.9	0.97
		3-GeV \bar{p}	5.160	2.514	2.05	0.15	14.92	63.0	0.32	63.5	0.54
U	2	880-MeV α	7.420	3.085	2.46	0.19	19.55	89.6	0.48	114.3	1.27
		700-MeV p	5.320	3.330	1.52.	0.09	18.94	85.4	0.22	52.9	1.20
		3-GeV \bar{p}	5.870	3.145	1.86	0.13	17.69	78.4	0.31	74.3	0.64

TABLE V. Summary of Mg²⁸ recoil results from the forward-backward experiments.^a

a Errors quoted in columns 4-6 are the standard deviation for replicate runs. Over-all errors are discussed in the Appendix.

			4		6	
Target	Number of det.	Bombarding particle and its energy	WF_{PF} (mg/cm ²)	WF_{PB} (mg/cm ²)	R_{0P} (mg/cm ²)	R_0/R_{0P}
Cu		3-GeV p	0.688	0.538	2.43	1.00
Au		880-MeV α 700-MeV p 3-GeV p	4.932 4.842 4.236	3.684 4.644 3.816	17.12 18.95 16.04	1.02 0.95 0.93
Ū		880-MeV α 3 -GeV p	4.080 4.800	3.710 4.290	15.5 18.1	1.26 0.98

TABLE VI. Summary of Mg²⁸ recoil results from the perpendicular experiments.⁸

a Over-all errors are discussed in the Appendix.

original nucleus into Na²⁴ and its complementary fragment with atomic charge $Z-11$ and atomic number $A-24$, where *Z* and *A* are the atomic charge and number of the target nucleus. For the purpose of a crude estimate we simply compute the energy of Coulombic repulsion of two tangent spheres with radii given by the expression $r = r_0 A^{1/3}$, where r_0 is taken as 1.45×10^{-13} cm.

The ratio of the experimentally measured recoil energy to this calculated E_{Coul} is a revealing quantity. If Na²⁴ is, in fact, produced as the result of the evaporation of many nucleons from an excited heavier nucleus, then the ratio should be low, say, $\langle 0.5$. A ratio approaching the value one would strongly suggest that the kinetic energy of the Na²⁴ comes from the Coulombic repulsion of large fragments.

The values of *E/Ecoui* are given in Fig. 9. All values are approximately equal to 0.7 indicating that a fissionlike division of the nucleus has probably occurred. The Coulombic energy has, to be sure, been estimated in a very approximate way but any correction to this quantity to allow for loss of cascade particles in advance of division of the nucleus, or for thermal expansion, or for shape deformations of the separating fragments will reduce the estimate of *Ecoui* and, hence, increase the ratio E/E_{Coul} . The large values of this ratio suggest that even in copper targets, and even at bombarding energies of 3 GeV, Na²⁴ and Mg²⁸ are not produced by a cascade followed by the evaporation of many nucleons, contrary to what one might have assumed. It is conceivable in the case of copper that the evaporation of heavier aggregates such as α particles could give rise to a larger value of E/E_{Coul} . The similar values of E/E_{Coul} suggest that the same mechanism may be responsible for fragment

FIG. 9. Ratio for Na²⁴ and Mg²⁸ of the intrinsic recoil energy to the Coulomb energy (see text) as a function of the target mass.

production in all target nuclei. This is a surprising result since, as indicated earlier, the excitation function data as interpreted by the conventional model show that at the bombarding energies used in this work more than 500 MeV of excitation are required in order to have "fragmentation." Such deposition energies are large enough to allow production of Na from Cu as the residue of a cascade evaporation process. This is emphasized by the fact that the total binding energy of copper is approximately 570 MeV and of silver approximately 920 MeV. If any sizeable fraction of the disintegrations leading to Na²⁴ production went by this nucleon evaporation mechanism the value of *E/Econi* should be smaller for Cu than for the other targets.⁴⁴

We turn now to a consideration of the values of *v,* defined as the velocity of the struck nucleus, and obtained from the recoil data with the aid of the Eqs. (4) and (5). Under the assumption that the progenitors of the fragments have mass values very nearly equal to the target mass, the average momenta, $\bar{P}_T = v_H \times A_{\text{target}}$, imparted to the struck nuclei in the beam direction have

⁴⁴ *Note added in proof.* Recent calculations by N. T. Porile (B.N.L.) indicate that it may be possible to account for the recoil energies of Na²⁴ produced from Cu by evaporation chains.

FIG. 11. Imparted velocity *v* (A) and fractional imparted
momentum \bar{P}_T/P_{CN} ;
(B) for Mg²⁸ as a function of target mass.

been calculated. These values are given in column 11 of Tables III and V. We have also computed the ratio \bar{P}_T/P_{CN} , where P_{CN} is the momentum of the hypothetical compound nucleus or equivalently, the momentum of the bombarding particle. These ratios are plotted in Figs. 10(B) and $11(B)$ for Na²⁴ and Mg²⁸, respectively. Because the progenitors of the fragments are expected to have somewhat less than the mass of the target nucleus when account is_ taken of the loss of particles in the initial cascade, \bar{P}_T is a slight overestimate of the actual momentum transferred. It is to be noted that the values of \bar{P}_T/P_{CN} are larger than one for 700-MeV protons and 880-MeV He ions incident on gold and uranium targets. This may be due to the overestimate of \bar{P}_T . It is also known that particular cascade events may result in struck nuclei with more forward momentum than is brought in by the bombarding particle if the internal motions of the target nucleons are considered. See, for example, Fig. 1 in an article by Porile.⁴⁵ For some unknown reason the process leading to Na²⁴ production may select out these particular cascade events. A third explanation, which we favor in our remarks below, is that the observed fragments are not produced by the two-stage process postulated in the conventional analysis so that the calculated quantities v_{11} and \bar{P}_T do not have a real meaning.

For the present, however, we continue to explore the implications of our data within the framework of the conventional two-stage mechanism to demonstrate a further major inconsistency. We call attention to the fact that the ratio, \bar{P}_T/P_{CN} , must have some relation to the ratio, E^*/E_{CN}^* ; in general, one would expect the forward component of momentum of the struck nucleus to increase as the amount of energy transferred to the struck nucleus increases. The quantitative interdependence of these quantities depends on the details of

45 N. Porile, Phys. Rev. **120,** 572 (1960).

the model of the prompt cascade process. Porile⁴⁵ examined the detailed cascade calculations of Metropolis *et aL^b* for targets of ruthenium, bismuth, and uranium bombarded with protons and was able to construct a plot, which we reproduce here as Fig. 12, showing the average deposited forward momentum as a function of the deposited energy, both expressed in units of the corresponding values for compound nucleus formation. It is seen that the deposited momentum increases almost linearly with increasing deposition energy, and as a first approximation, the relationship between P_F/P_{CN} and E^*/E_{CN} ^{*} can be considered to be independent of the bombarding energy and target material. There would seem to be no reason why this should not be true also for the targets used in our study.

If we now re-examine our values of P_T^*/P_{CN} we note that there is a strong increase in this value with increase in target mass number, which implies an increase in excitation energy *E** with increase in target mass number. But in our discussion of the excitation function results, we were led to the conclusion that *E** is independent of the target mass. This is a pronounced inconsistency and it is hard to see how more refined calculations of the cascade and evaporation steps can remove it. It may be that the cascade results which lead to fragmentation are so atypical that it is incorrect to compare results with the predictions of average prompt cascade calculations. Alternatively, the production of such fragments as Na²⁴ may go by a mechanism alien to the cascade-evaporation model.

It is noteworthy that the measurements imply a strong similarity in the average processes leading to Na²⁴ or Mg²⁸ production from all targets: Excitation functions are almost proportional; the values of v_{II} and *E/Ecoui* are only slightly dependent on target mass for each incident energy. The prompt cascade calculations⁵ lead one to expect much more variation in these quantities.

IV. FRAGMENTATION MECHANISMS

A. Experimental Findings Requiring Explanation

The difficulties and inconsistencies cited above lead us to abandon the prompt-cascade-slow-de-excitation description of the mechanism responsible for production of

Na²⁴ and Mg²⁸ in proton-induced reactions. We believe that this conclusion must also be extended to the helium-ion reactions. Let us summarize some of the main features for which an adequate reaction model must account.

(1) There is negligible production of such products as Na²⁴ and Mg²⁸ until the energy of the bombarding particle exceeds a few hundred MeV. Excitation functions rise steeply above this high "threshold" until bombarding energies exceed 1 GeV, above which energy there is a pronounced leveling off in yield.

(2) The excitation functions for Na^{24} and Mg^{28} have the same shape whether protons or helium ions are used as bombarding particles, but the absolute cross sections run a factor of approximately 2 higher when helium ions are used.

(3) The ratio of the yield of Mg^{28} to that of Na^{24} is substantially less than one and for a given target remains remarkably constant when the helium-ion energy is varied or when protons are substituted for helium ions as the bombarding particle. See Table I.

(4) The yield of these products goes through a minimum as a function of target atomic number as shown in Figs. 5 and 6. This is true for the helium-ion bombardments as well as the proton bombardments.

(5) Na^{24} and Mg^{28} have very substantial recoil energies in all cases studied. These energies are roughly equal to that which they would receive by Coulombic repulsion in a fission-like division of the target nucleus into two large pieces. It is especially noteworthy that high kinetic energies are observed for these products even in the case of copper targets.

(6) The forward-backward asymmetry of the Na^{24} and Mg²⁸ recoils are apparently not consistent with a process having an angular distribution symmetric about 90 deg.

(7) If the fragments observed in emulsion studies are produced in the same type of events as those which produce Na^{24} or Mg^{28} , then there is a whole series of their properties which must be accounted for. Among them we may mention such features as their anisotropic emission (strong forward peaking), their unexpectedly high rate of multiple emission, and the favoring of fragments of low excitation near the line of beta stability. These and other features are discussed fully in the review paper of Perfilov, Lozhkin, and Shamov.¹⁰

B. Nature of the Fragmentation Mechanism

Several authors have made qualitative proposals for the nature of the fragmentation mechanism.^{25,46-50} These

proposals have two common features: (a) The process involves large transfers of energy from the projectile to the constituents of the target nucleus; (b) the process is fast with respect to the nuclear rotation period. The cross-section data [item (1) above] lead to the feature (a). The forward-backward asymmetries observed in this study [item (6) above] and other work [item (7) above] point to feature (b).

Wolfgang and co-authors²⁵ postulated that fragment production occurred as the result of the near simultaneous breaking of many nucleon-nucleon bonds in a restricted volume of the nucleus during the prompt cascade initiated in heavy targets by protons of 0.6- to 3-GeV energy. The disturbance of the internal nuclear structure is believed to be so large that the large fragments are emitted or split off before any sort of equilibrium equipartition of energy can occur. Surface tension and Coulomb repulsion forces as well as momentum imparted by the knock-on cascade tend to separate clumps of nucleons from each other. These authors postulate that one important contribution to the development of the prompt cascade is the production and reabsorption of π mesons.

Lozhkin⁴⁹ also emphasizes the importance of a complex fast prompt cascade in a restricted region of the nucleus although he does not include the production and reabsorption of mesons as a significant element of the development of the cascade. He speaks of local volume and surface distortions which become increased by the interplay of surface tension and Coulombic repulsion forces.

Faissner and Schneider⁵⁰ also describe in a qualitative way a mechanism for induction of fast fission modes in heavy-element targets bombarded with 600-MeV protons. Their mechanism is qualitatively rather similar to the above.

C. The Helium-Ion Results and Fragmentation

We conclude from our results \lceil items (2) and (3) above] that the cascade initiated by a high-energy helium ion is remarkably similar to that initiated by a proton of the same energy.⁵¹ In the absence of detailed information on the elastic and inelastic scattering of high-energy helium ions from single nucleons it is difficult to decide how to set up a prompt cascade calculation for the interaction of a high-energy helium ion with a complex nucleus. We think it unlikely that mesons are produced in nearly the same intensity by helium ions compared to protons. Hence, we assert that meson production and reabsorption cannot play a significant role in fragment production in our experiments

⁴⁶ P. Kruger and N. Sugarman, Phys. Rev. 99, 1459 (1955).
⁴⁷ D. I. Blokhintsev, Zh. Eksperim. i Teor. Fiz. 33, 1295 (1957)
[⁴⁷ anslation: Soviet Phys.—JETP 6, 995 (1958)].
⁴⁸ A. E. Glassgold, W. Heckrotte and K. M Cosmic Rays, 1961.

⁵¹ Additional evidence in support of this point comes from an unpublished study by Korteling and Hyde in which the excitation functions of a variety of products were studied in the interaction of high energy protons and helium ions with niobium targets over the bombarding energy range 240-880 MeV. The results of the proton-induced reactions were remarkably similar to those for helium-ion-induced reactions.

done with helium ions. We cannot prove that meson effects are negligible in the proton bombardments for it may be that the participation of the four nucleons of the alpha particle in the prompt cascade gives rise to a greater development of this cascade which closely compensates for the absence of meson contributions to the cascade. However, we think it is strange that the compensation should be so exact that the cross-section trends and the recoil properties should be so similar in all the targets and all the bombardment energies used in our experiments. Hence, we tend toward the view that meson effects are not a major contributor to the production of fragments even in proton-induced fragmentation.

We agree with the cited authors that the action of surface and Coulombic forces during and after the prompt cascade is important in determining the final products observed in radiochemical experiments, but we see no way at present to set up a calculation of the action of these forces.

The presence of a minimum in the yield of Na²⁴ and Mg²⁸ plotted against the target atomic number is a very interesting feature. Caretto, Hudis, and Friedlander²⁶ accounted for such a minimum in the results obtained in the bombardment of a series of targets with protons in the energy range 1-6 GeV by postulating the operation of two formation mechanisms. In the lighter targets the observed products were thought to be evaporation residues left after de-excitation by emission of many neutrons and protons. In the heavier targets they were believed to be fragmentation products. Our result of a high kinetic energy for Na²⁴ formed in copper targets suggests that in this light target it also is a fragmentation product. If it is true that a minimum occurs for a single type of process, the explanation for this may possibly lie in the energetic requirements of the distortions of various target nuclei which lead to the production of specific nuclei by a fission-like splitting. From liquid-drop calculations, particularly those of Cohen and Swiatecki,⁵² it is known that as a function of charge and mass there are marked changes in the absolute amounts of energy required in particular types of distortions as well as marked changes in the favoring of certain types of distortions over others. Big changes in the energetic requirements may influence the outcome of any process in which the development of a distortion in the nucleus by a fast or slow process is decisive even if energy equilibration between all possible distortion modes is not achieved.

This discussion is necessarily qualitative since neither our results nor previous results provide a clear picture of the nature of fragmentation. Although the development of a complex knock-on cascade may be essential as a first step to fragment production we have no reaction model to describe the ultimate behavior of the struck

nucleus on its way to division into fragments or fragment precursors.

Our main contributions are these. We have shown that the yields and recoil properties of products such as Na²⁴ and Mg²⁸ cannot be explained by the conventional reaction model of a fast cascade followed by a slow process (a process resulting in a symmetric angular distribution). We have also shown that the yields and recoil properties in the bombardments done with helium ions and protons are remarkably similar. This raises interesting questions about the high-energy cascade induced by helium ions.

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APPENDIXES

A. Radiochemical Procedures

The chemical procedures were adaptations of standard radiochemical methods.53,54 The metal foils were dissolved with appropriate acids and the plastic foils destroyed with a hot mixture of sulfuric and nitric acids in the presence of 10 mg of Na carrier and 5 mg of Mg carrier. The sulfuric-nitric acid mixture was evaporated to dryness and the residue dissolved in distilled water. The target materials were removed by precipitation of copper sulfide, silver chloride, or uranium tetroxide.⁵⁵ or, in the case of gold targets, by extraction of gold with ethyl acetate.

After this removal of the bulk of the target material the solution volume was adjusted to 10 ml. Iron hydroxide was precipitated once in the presence of ammonium chloride to scavenge out unwanted hydroxide-insoluble contaminants, and then copper, antimony, and nickel sulfides were precipitated. The excess hydrogen sulfide was removed and another precipitation of iron hydroxide was performed. Magnesium was precipitated with 8 hydroxyquinoline and the solution kept for sodium analysis. The magnesium hydroxyquinolate was destroyed. The hydroxide and sulfide precipitations were repeated. Calcium, strontium, and barium oxalates

⁵² S. Cohen and W. J. Swiatecki, Ann. Phys. (N. Y.) 19, 67 (1962) and University of California Lawrence Radiation Laboratory Report, UCRL-10450, 1962 (unpublished).

⁵³ J. R. Grover, Phys. Rev. **126,** 1540 (1962) and University of California Radiation Laboratory Report UCRL-3932, 1957 (unpublished).

⁶⁴ Los Alamos Scientific Laboratory Report, LA-1721, 1954 (unpublished).

⁶⁵ *Analytical Chemistry of the Manhattan Project* (McGraw-Hill Book Company, Inc., New York, 1950), p. 48.

were twice precipitated and the final supernatant liquid was evaporated to dryness. The residue was taken up in water and strontium sulphate was precipitated *(pH=6).* Iron hydroxide was precipitated once again and removed by filtration. Magnesium was precipitated with 8-hydroxyquinoline, filtered, dried at 110°C, weighed, and mounted for counting.

The solution, containing the sodium fraction, was treated with a mixture of benzene and 1 butanol to remove excess hydroxyquinoline (and possibly quinolinates of impurities). The organic solution was discarded and the aqueous solution was evaporated to dryness. The residue was dissolved in water. Sodium was precipitated as sodium magnesium uranyl acetate and converted to sodium chloride with hot 1-butanol saturated with dry HC1. This step was repeated. The sodium chloride precipitate was treated with concentrated perchloric acid, 5 mg of potassium chloride was added and the solution evaporated to dryness. Sodium perchlorate was extracted with 1 butanol and converted into sodium chloride with 1 butanol saturated with hydrogen chloride. The sodium chloride was filtered, dried at 110° weighed, and mounted. The chemical vield of Na^{24} or Mg^{28} was assumed equal to the percentage recovery of the 10 mg of inert sodium or 5 mg of magnesium added in the beginning.

The filtration apparatus has been described by Blann.⁵⁶ In part of this work, glass filter pads were used to minimize weighing errors due to absorption of moisture.

B. Mounting of Samples and Determination of Their Radioactivity

Samples were mounted on aluminum plates (350 mg/cm²) and covered with 0.1-mil Pliofilm. The Na²⁴ and Mg²⁸ activity was measured by placing the sample in a reproducible geometry 0.9 cm from an end-window methane-flow proportional counter of 2.54-cm diam. Measurements were repeated in a regular rotative sequence with a series of counters in order to minimize variations in efficiency and background. This rotation of samples was particularly important in the samples from the recoil catcher foils, particularly the backward

catcher foils. Standard checks of counter performance were made daily.

The influence of sample thickness upon counting efficiency was determined by comparison of the counting rates of Na²⁴ in a stack of aluminum foils of different thicknesses bombarded with protons in a special irradiation. The relative counting efficiencies of Na^{24} and Mg^{28} was taken from Bayhurst and Prestwood.⁵⁷

In the samples obtained from the recoil experiments it was necessary to evaluate error due to activation of impurities in the catcher foils. The Na²⁴ produced in blank foils was always measurable and corresponded to a small percentage of the total Na²⁴ activity found in the catchers. In the Mg²⁸ experiments, on the other hand, the activities were much lower and background activity due to activation was not measurable. However, since the activity of the Mg²⁸ samples was very low, especially in the backward recoil catcher, activations of the order of 1 count/min could have introduced errors up to 10% .

In every Na²⁴ sample there was a prominent 15-h period which could easily be resolved from the background. Chemical yields were practically constant for all foils from a given experiment. The Mg²⁸ activities were lower, the chemical yields somewhat variable, and the possible contribution from contaminants was higher. The uniformity in thickness of the copper, gold, and uranium targets was so high that there was negligible error from this source. Errors from thickness nonuniformity in the silver foils may have reached 5% . Errors caused by oxide film on the uranium are considered to be small. The over-all error of the measurement (at the 95% confidence level) of the product, $W \times F$, in the recoil experiments is considered to be of the order of 5% for Na²⁴ produced in copper, gold, and uranium, and of the order of 10% in silver. The Mg²⁸ errors are probably about twice as large.

C. Effects of Approximations Made in Calculating Ranges of Fragments and Velocities of Their Progenitors

A. The Effect of Neglecting η_1

Equations (1) , (2) , and (3) given in the text can be rewritten in the following forms:

$$
\frac{F_F}{F_B} = \frac{\left[1+\eta_{11}\frac{2}{3}(N+2)+\eta_{11}\frac{(N+1)^2}{4}\right]+\eta_{1}\frac{N^2-1}{8}+\frac{b}{a}\left[\frac{1}{6}+\eta_{11}\frac{8(N-1)}{45}+\eta_{11}\frac{N^2-N-4}{12}+\eta_{1}\frac{N-1}{12}\right]}{\left[1-\eta_{11}\frac{2}{3}(N+2)+\eta_{11}\frac{(N+1)^2}{4}\right]+\eta_{1}\frac{N^2-1}{8}+\frac{b}{a}\left[\frac{1}{6}-\eta_{11}\frac{8(N-1)}{45}+\eta_{11}\frac{N^2-N-4}{12}+\eta_{1}\frac{N-1}{12}\right]},
$$
\n
$$
W(F_F-F_B) = R_0\eta_{11}\left[\frac{N+2}{3}+\frac{b}{a}\frac{4(N-1)}{45}\right],
$$
\n(8)

66 H. M. Blann, University of California, Lawrence Radiation Laboratory Report UCRL-9190, 1960 (unpublished).

⁵⁷ B. P. Bayhurst and R. J. Prestwood, Nucleonics 17, 82 (1959).

and

$$
4WF_P = R_0P \left[1 + \eta_{11}^2 \frac{(N-1)(N+1)}{8} \right]
$$

+
$$
R_0P \left\{ \eta_1^2 \frac{(N+1)(3N+1)}{16} + \frac{1}{6} \frac{b}{a} \left[-\frac{1}{2} + \eta_1^2 \frac{(7-3N)(N+1)}{16} + \eta_{11}^2 \frac{(N-1)(N-5)}{8} \right] \right\}.
$$
 (9)

It will be seen in Appendix D that *N* takes the value 1.5 in Cu, Ag, Au, and U. In this case Eq. (7) becomes

$$
\frac{F_F}{F_B} = \frac{1+2.33\eta_{11}+1.56\eta_{11}^2[1+0.1\eta_{12}^2/\eta_{11}^2]+0.04(b/a)\eta_{12}^2+(b/a)(\cdots)}{1-2.33\eta_{11}+1.56\eta_{11}^2[1+0.1\eta_{12}^2/\eta_{11}^2]+0.04(b/a)\eta_{12}^2+(b/a)(\cdots)}.
$$
\n(10)

By inspection of Eq. (10) we decide that η_1/η_{11} must be much larger than 1 in order to introduce appreciable errors into the estimate of $\eta_{\rm II}$ if F_F/F_B is not extremely large. In order to get an estimate of the relative magnitudes of η_1 and η_{11} we refer to the information which Porile⁴⁵ extracted on this point by an examination of results of Monte Carlo calculations of the nucleonnucleon cascade. He found that for large momentum transfers (such as are important in our experiments) the ratio η_1/η_1 is indeed much less than unity. Thus, the neglect of η_{\perp} is justified.

Since $R_0 \eta_{11}$ is independent of η_1 [Eq. (5)], errors due to the neglect of η_1 are equally introduced in η_{II} and R_0 . However, $W(F_F-F_B) \propto R_{0} \eta_{11} \propto V^{1/2} v_{11}$ (for $N=1.5$) and, therefore, v_{11} is practically independent of η_1 .

B. The Effect of Neglecting b/a

Possible effects of nonisotropic distribution of the products in the center-of-mass system of the progenitor are expressed by the term in b/a in Eq. (8) which for $N=1.5$ reduces to

$$
W(F_F-F_B) = k^{2/3} (1.16 + 0.044b/a) R_0^{1/3} v_{11}.
$$

For values of *b/a* smaller than 0.5 the influence of *b/a* on the expression enclosed in parentheses is negligible. The experimental data from \hat{WF}_p and $\hat{W}(F_F+\hat{F}_B)$ measurements indicate that b/a is always less than 0.5. Thus, the calculation of $v_{\rm H}$ is only affected through the influence of b/a on R_0 .

C. The Effect of a Distribution of Values of v or V

The assumption that *v* and *V* are unique is not expected to be strictly correct. This introduces certain errors in the analysis. The measured quantities are the average quantities \overline{F}_F and \overline{F}_B . For Na²⁴ and Mg²⁸ in all targets $N=1.5$. For $N=1.5$ Eqs. (4) and (5) can be written

and

$$
\left\langle W(F_{F}-F_{B})\right\rangle \propto\left\langle V^{N-1}v\right\rangle
$$

 $\langle W(F_F+F_B) \rangle \propto \langle V^N[1+1.562(v/V)^2] \rangle$

The values of $1.562(v/V)^2$ are usually much smaller

than 1. Thus,

 $\langle V^N[1+1.562(v/V)^2]\rangle \cong \langle V^N \rangle[1+1.562(v/V)^2]$

and since N is close to 1,

 $\langle V^N \rangle^{1/N}$ \cong $\langle V^N \rangle$

Therefore, $\langle W(F_F - F_B) \rangle \cong k' \langle V \rangle^N \langle v / V \rangle$, and $\langle v / V \rangle$ and *(V)* may be obtained. The estimate of *(v)* from the product $\langle V \rangle$ and $\langle v / V \rangle$ is more accurate than either factor separately. It is estimated that *(V)* and *(v)* can be obtained with an error smaller than 10%. Errors of this magnitude do not invalidate the arguments presented in the text. This is especially true because similar errors are probably introduced for all targets and, thus, the relative errors are expected to be less than the absolute errors.

D. The Effect of Scattering

A charged particle going through matter loses its energy by electronic and atomic collisions. Electronic collisions are responsible for the losses of energy suffered by fast-moving recoils. On the other hand, slow recoils transfer their energy to the nuclei of the stopping medium. When the masses of the atoms in the medium are comparable to or larger than the mass of the recoil, the recoiling atoms suffer large deflections in almost every collision. As a result of these large deflections more fragments scatter out of the target material than back from the plastic catchers,⁵⁸ increasing the apparent recoil ranges. This nuclear scattering effect becomes important for speeds of the recoils lower than a critical speed V_c given by⁵⁹

$$
V_c^2 = 2Z_1Z_2e^2(Z_1^{2/3} + Z_2^{2/3})^{1/2}/\mu a_0,
$$

where Z_1 and Z_2 are the atomic numbers of the stopper and stopping nuclei, μ the reduced mass of the system, *e* the electronic charge, and *ao* the first Bohr radius $(0.528 \times 10^{-8} \text{ cm})$. Niday⁵⁸ has measured this effect for thick-target recoil studies of the fission products in uranium. He found the apparent ranges of the fission

⁵⁸ J. B. Niday, Phys. Rev. **121,** 1471 (1961). 69 N. Bohr, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. 18, No. 8 (1958).

products caught in Al to be larger by approximately 3% for heavy-fission products and 5% for light-fission products than the corresponding ranges of the fission products caught in lead. The V_c 's for Na²⁴ (and Mg²⁸) fragments are lower than the *Vc's* for the fission products. Furthermore, the ratios of the initial speed of the fragments to V_{c} are larger for Na²⁴ (and Mg²⁸) than the corresponding ratios for the fission fragments. Therefore, the nuclear-scattering effects are expected to be relatively less important for the Na²⁴ and Mg²⁸ fragments observed in this study than for the fission products. Furthermore, the values of *v* are affected only by the difference in the scattering effect for the backward and forward fractions of recoiling nuclei. Therefore, we consider the corrections due to scattering effects to be of second order and they have been neglected.

D. Range-Energy Relationships

The required range-energy relationships needed in our work for Na²⁴ and Mg²⁸ stopped in copper, silver, gold, and uranium were not available in the published literature and could not be easily obtained from theory. Consequently, we derived the needed relationships indirectly in the following way.

Heckman and co-workers⁶⁰ have measured the ranges in nuclear emulsions of nuclei of C, N, O, Ne, and Ar accelerated to energies of 0.5 to 10 MeV per atomic mass unit. They used the range, $\lambda(\beta)$, of a proton with velocity $\beta = v/c$ as their standard and discussed the observed ranges of the complex particles in terms of the corresponding range, $(M/Z^2) \lambda(\beta)$ (expected if no neutralization of charge occurred) and in terms of a range extension term, R_{ext} (β), which corrected for the pickup of charge by the fragment as it loses energy. The range extension is defined by

$$
R_{\rm ext}(\beta) = R(\beta) - (M/Z^2) \lambda(\beta) ,
$$

60 H. H. Heckman, B. L. Perkins, W. G. Simon, F. M. Smith, and W. H. Barkas, Phys. Rev. **117,** 544 (1960).

where $R(\beta)$ is the observed range. Heckman *et al.*⁶⁰ found that a plot of $R_{ext}/MZ^{2/3}$ versus the velocity of the ions, expressed in units of the K -electron velocity, gave a universal curve. We evaluated the ranges of Na²⁴ and Mg²⁸ in emulsion by use of this universal curve and the above relation. In the energy interval 0.4 to 3.0 MeV/ amu, the ranges are given by $R=0.95(E/A)^{0.94}$, expressed in mg/cm² . This range-energy relationship should be fairly accurate, since in this same energy interval the ranges of all complex particles are, for a given speed, the same within 20% .

The relative stopping powers of the nuclear emulsions and the target materials used in this work, for ions of a given speed, were determined by the ratios of the ranges of α particles or protons of the same velocity in nuclear emulsions and in the target material. This procedure is not absolutely correct, but it is a reasonable approximation. The ranges in emulsion of α particles and protons were taken from Barkas *et al.⁶¹* and Heckman *et* al.,⁶³ respectively. The ranges of α particles and protons in Cu, Ag, and Au have been taken from Whaling.⁶² The ranges of the fragments in uranium were assumed equal to the ranges of the fragments in gold. While there is published evidence^{57,63,64} that this assumption is not strictly correct, the errors introduced by this assumption in the calculated recoil energies of the fragments produced in uranium should not exceed 10%. The range-energy relationships obtained for both Na²⁴ and Mg²⁸ are of the form $R = k(E/A)^{0.76}$, where k is a constant. With *R* expressed in mg/cm² and E/A in MeV/amu, the constant *k* takes the values of 4.5 for Cu, 5.8 for Ag, and 8.1 for Au and U.

⁶i W. H. Barkas, P. H. Barret, P. Ctier, H. Heckman, F. M. Smith, and H. K. Ticho, Nuovo Cimento 8, 185 (1958).

⁶² YV. Whaling, in *Handbuch der Physik,* edited by S. Fliigge (Springer-Verlag, Berlin, 1958), Vol. 34, p. 193.

⁶³ J. M. Alexander and M. F. Gazdik, Phys. Rev. **120,** 874 (1960).

⁶⁴ A. Garin and H. Faraggi, J. Phys. Radium 19, 76 (1958).