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Gordon, Glen E.

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REACTIONS INDUCED BY HEAVY IONS

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UNIVERSITY OF CALIFORNIA
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INDUCED BY HEAVY IONS

Glen E. Gordon
(Thesis)

May 1960

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Glen E. Gordon

Lawrence Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

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ABSTRACT

Several features of fission and spallation reactions proceeding through astatine compound nuclei formed by carbon-ion and nitrogen-ion bombardment have been investigated. The kinetic-energy spectra of the fission fragments were observed at various angles to the beam over a range of bombarding energies by use of two types of detectors, gas scintillation chambers and diffused p-n junctions. Cross sections for neutron-evaporation reactions were determined by radiochemical measurement of the production of astatine isotopes.

Analysis of the fission-fragment angular distributions according to the models by Halpern and Strutinski and by Griffin, together with the dependence of the fission and spallation cross sections on bombarding energy, suggests that fission is frequently preceded by evaporation of neutrons and charged particles. This result is explained on the basis of increasing probability for charged-particle emission with excitation energy and hindrance of neutron evaporation at low energies due to angular-momentum and level-density effects. The latter argument is also used to explain discrepancies between experimental and theoretical shapes of the excitation functions for neutron-evaporation reactions. Evidence has also been found that, in the astatine region, a larger total kinetic energy release is associated with symmetric fission than with the asymmetric modes. This is a reversal of the trend found in heavier elements.

In order to obtain the data presented in the main body of the thesis, several supplementary investigations were necessary. These are discussed in the appendices.

FISSION AND SPALLATION IN NUCLEAR REACTIONS INDUCED BY HEAVY IONS

Glen E. Gordon

Lawrence Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

May 1960

I. INTRODUCTION

The work presented here is a study of fission and spallation reactions induced by heavy-ion bombardments in which the compound nuclei formed are in the astatine region. A number of quantities pertaining to fission have been measured, as well as some excitation functions for spallation reactions.

Nuclear fission was discovered by Hahn and Strassman in 1938.¹ Since then, many attempts have been made to interpret the fission process theoretically. Most of the treatments have been performed in the framework of the liquid-drop model, originally proposed by Bohr and Wheeler² and Frenkel.³ According to this theory, the nucleus is pictured as a uniformly charged liquid drop. Fission occurs if the liquid drop is distorted beyond the point where further distortion decreases the Coulomb energy faster than it increases the surface energy of the drop. A fundamental quantity that evolves from this model is Z^2/A . Although some correlations of relative fissionabilities^{4,5} and spontaneous-fission half lives^{6,7} with Z^2/A have been moderately successful, it is not clear that all the properties of the fission process can be explained by the liquid-drop model.

Probably that property of the fission process most difficult to explain is the distribution of masses of the fission fragments. At low excitation energies, in the actinide elements, asymmetric fission events are much more probable than symmetric events.^{8,9,10} As the energy is raised, symmetric division becomes more probable. Various explanations for the large probability for asymmetric division at low energies have been

advanced.¹¹⁻¹⁵ Some of these theoretical treatments have yielded good agreement with the low-energy experimental results, but it would appear difficult for them to explain the change in mass-yield distributions with increasing energy. The situation is further complicated by the results of Fairhall et al., who find that at the lowest energies at which fission is experimentally observable in elements below actinium, the mass-yield curves consist of single, narrow peaks centered at approximately half the mass of the fissioning nucleus, indicating that symmetric fission is highly favored.¹⁶ A very striking result is obtained when Ra^{226} is bombarded with 11-Mev protons. In that case, the mass-yield curve exhibits three peaks, indicating roughly equal probabilities for symmetric and asymmetric division.

Much of the information on relative fissionabilities in the heavy element region has been obtained by measurements of spallation yields in reactions induced by charged particles. This method, originally devised by Glass and co-workers¹⁷ and refined by Vandenbosch et al.,¹⁸ involves determination of the cross sections for neutron-evaporation reactions and comparison of these experimental results with cross-sections calculated according to the Jackson model.¹⁹ Since the Jackson calculations have been successfully fitted to the experimental cross-sections in regions where fission is not a competing reaction, differences between calculated and experimental neutron-evaporation cross sections in the heavy-element region have been attributed to fission competition.

The results of many of these studies in the heavy-element region are consistent with the assumption that Γ_f/Γ_n (the ratio of level width for fission to level width for neutron emission) for a given nucleus is independent of energy.^{4,17,18,20} However, in elements with $Z < 90$, there is considerable evidence that Γ_f/Γ_n is a strongly increasing function of excitation energy.^{8,16,21} Fairhall, Jensen, and Neuzil suggest that these results, and the mass-yield data discussed above, can be explained by the assumptions that (a) symmetric and asymmetric fission are two distinct processes; (b) the probability for symmetric fission has a strong dependence on Z , but little A dependence; (c) the probability for symmetric fission

(relative to neutron emission) increases up to some limiting value with increasing bombarding energy; and (d) at energies above the neutron binding energy plus the fission barrier, the probability for asymmetric fission is a decreasing function of excitation energy.¹⁶

In addition to the probability of fission, several other quantities related to fission have been of interest. Terrell has successfully correlated the average total fragment kinetic energy release with $Z^2/A^{1/3}$ of the fissioning nucleus.²² This result implies that fragment kinetic energies arise from mutual Coulombic repulsion of the two fragments. Quantitatively, the kinetic energies are about 25% lower than expected on the basis of such a model. This is presumably due to distortion of the fragments from sphericity at the time of scission. This model, however, has not successfully explained the variation of total kinetic energy release with the ratio of fragment masses for a given fissioning nucleus (see Section IV).

In 1955, Bohr proposed a model which explained the then existing data on fragment angular distributions resulting from photon- and neutron-induced fission.²³ Since then, Halpern and Strutinski²⁴ and, independently, Griffin²⁵ have extended this model quantitatively to explain more recent angular-distribution data from neutron- and charged-particle-induced fission. These theories are discussed in detail in Section IV.

Heavy-ion-induced fission and spallation reactions should be particularly interesting for a number of reasons. For one thing, the particle energies obtainable from the Berkeley heavy-ion linear accelerator (hereinafter referred to as the "Hilac") make possible the study of compound nuclei at very high excitation energies (for example, approx. 100 Mev for full-energy C^{12} bombardments). The calculations of Ericson²⁶ indicate that the compound-nucleus model, originally proposed by Bohr,²⁷ should retain much of its validity at these high energies when formed by heavy-ion bombardment. This is in contrast to reactions induced by smaller particles, say protons, in which interpretation of the results is made more difficult by direct particle-particle interactions that may take place during the initial phases of a nuclear reaction.²⁸

Probably the most interesting feature of heavy-ion reactions is the large amount of angular momentum that may be imparted to the compound nucleus by the incoming beam particle.²⁹ Pik-Pichak's calculations predict that the effective barrier heights for fission are reduced as a result of large angular momenta.³⁰ Such an effect would increase Γ_f/Γ_n relative to its value at low angular momenta. There is another effect which also tends to produce the same trend. The calculations of Ericson indicate that high-spin states are rather improbable in nuclei excited only to energies of the order of the neutron binding energy or lower.³¹ Thus the probability for neutron emission from high spin levels to residual nuclei at low excitation energies is decreased relative to its value when the evaporation proceeds from a low-spin state. As a result of high angular momentum, one therefore expects (a) fission to compete more successfully with evaporation of small particles, such as neutrons, protons, etc., and (b) neutron evaporation to compete less successfully with fission and charged-particle emission than at low angular momenta. Huizenga has discussed some results which are in agreement with those predictions.²¹

In this work, several of the quantities related to heavy-ion-induced fission have been studied. In order to do this, fission fragments were observed with two types of detectors, gas-scintillation and solid-state detectors. By observing the spectra of fragment kinetic energies at various angles to the beam it was possible to obtain information about fragment angular distributions, total kinetic energy release, fission cross sections, and momentum transfer by the bombarding particle to the compound system. Also, some cross sections for neutron evaporation resulting from the bombardment of platinum with N^{14} ions have been determined.

There were a number of reasons for concentrating these studies on compound nuclei in the astatine region. The astatine isotopes which result from neutron-evaporation reactions decay by alpha-particle emission with half lives which are convenient for observation.³² However, this advantage is somewhat negated by uncertainties in the alpha/EC branching ratios and the possible existence of unobserved isomers, particularly for the odd-odd isotopes. The compound nuclei that are formed are near the region in which Fairhall

et al. have observed the drastic changes in mass-yield distributions.¹⁶ Also, it was expected that fission would not predominate over other reactions as much as it does in the heavy-element region. There are many data available for possible correlations in this region. Besides the fission results already mentioned above, Polikanov and Druin have measured many heavy-ion-induced fission cross sections in this region.³³ Baraboshkin, Karamyan, and Flerov have determined several (N^{14}, xn) reaction cross sections on gold,³⁴ and, concurrent with this research, Latimer and Thomas studied the (C^{12}, xn) reactions on gold.³⁵ In addition, considerable data on the spallation excitation functions of $Bi^{209} + He^4$ are available.^{5,36} Tarantin et al. have investigated the fragment mass-yield distribution from fission of gold with 115-Mev N^{14} particles.³⁷ Preliminary work on the spallation cross sections in the $Pt + N^{14}$ system has been reported by Burcham and Haywood.³⁸

The experimental procedures and results are discussed in Section II and III, and the analysis of the results is discussed in Section IV. Some supplementary investigations are described in the appendices.

II. FISSION COUNTING EXPERIMENTS

A. Experimental Procedures

1. Gas Scintillation Method

a. Introduction

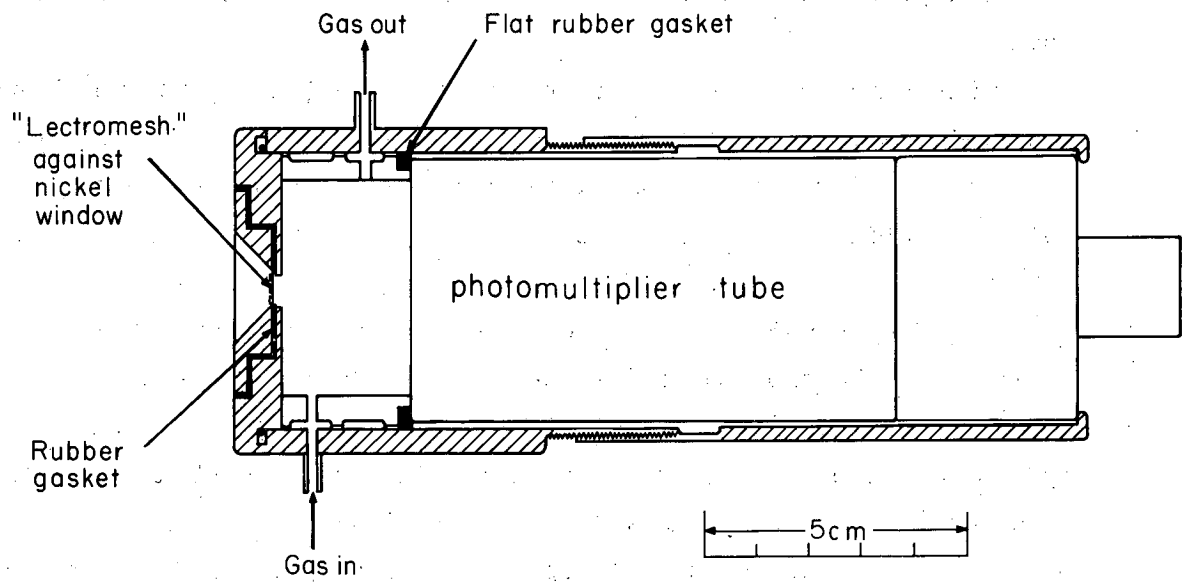
A fairly complete survey of the field of gas scintillation counting has been given by Sayres and Wu.³⁹ This technique has been used by Nobles and Leachman for observation of the kinetic energy spectra of fragments from the fission of Ra^{226} by neutrons.⁴⁰

The gas scintillation method has two important advantages over techniques previously used for counting and analyzing the energy of fragments from beam-induced fissions. First, the pulses obtained have fast rise times (approx 10^{-8} sec).⁴¹ This was quite important in the fragment-counting experiments because, at angles near the beam direction or at low bombarding energies, the ratio of scattered beam particles and light reaction products to fission fragments was quite large. In these cases it was necessary to withstand large counting rates of low-energy pulses in order to obtain good statistics on the number of fission-fragment counts in a reasonable length of time. Under these conditions, a slower counter would have made the "pile-up" of small pulses more serious. Unfortunately, no fast electronic system for amplification and discrimination of the pulses was available during the work, so that much of this advantage was lost.

The other chief advantage of this technique is that the scintillating material is of low density, and therefore, rather insensitive to the beta and gamma rays and neutrons associated with the heavy-ion beam. It was possible to make the counter just long enough to stop the densely ionizing fission fragments while absorbing only a small fraction of the energy of the scattered beam particles.

b. Construction and Characteristics of the Counter

A schematic diagram of the gas scintillation chamber is shown in Fig. 1. The fragments entered the chamber through a 0.03-mil Ni foil window, 1/4 in. in diameter, supported against the vacuum by a 49%-transmission "Electromesh" grid.⁴² The photomultiplier tube (DuMont 6292)



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Fig. 1. Schematic diagram of the gas scintillation counter.

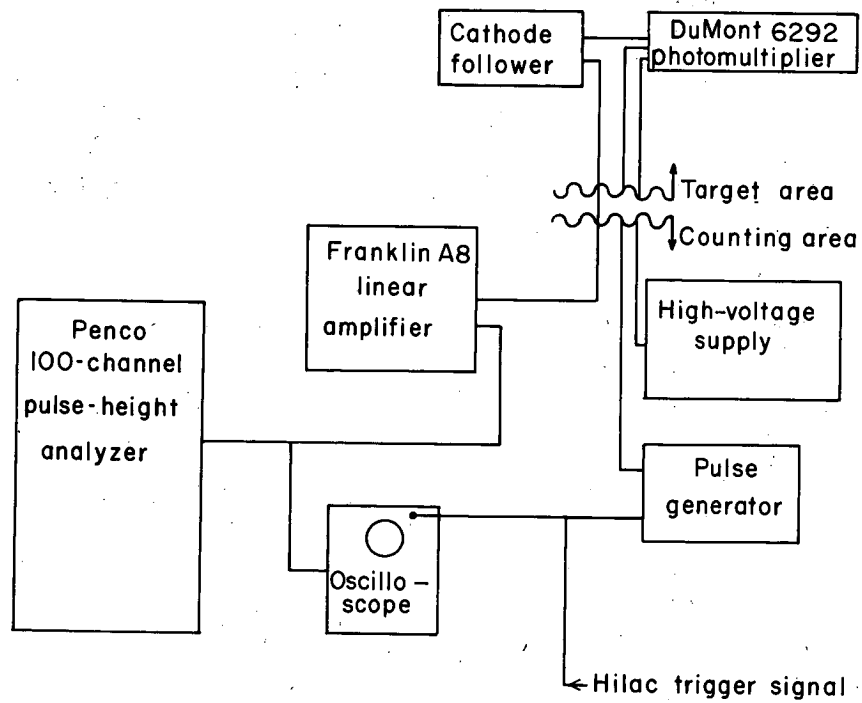
was seated on a flat rubber gasket on one end of the cylindrical gas chamber. The scintillating gas was argon, which was flushed through the chamber at atmospheric pressure. At the start of each series of experiments, gas was flushed through the chamber for at least 30 min. in order for the impurity concentration to decrease to an equilibrium value. It was possible to determine that stability had been attained when the pulse heights of successive Cf^{252} spontaneous-fission calibrations were the same.

The chamber walls were coated with Tygon (T10) paint which served as the reflector. Its performance was equal to that of the more commonly used MgO , and it was mechanically more stable. The "wave-length shifter," diphenyl stilbene, was sublimed onto the chamber walls and the face of the photomultiplier tube.

A block diagram of the electronic system is shown in Fig. 2. High voltage on the photomultiplier tube was normally maintained at 1000 to 1100 volts. After amplification by the linear amplifier, pulses from the photomultiplier were sorted and stored in the Penco (Pacific Electro-Nuclear Co. 100-channel analyzer, Model PA-4).

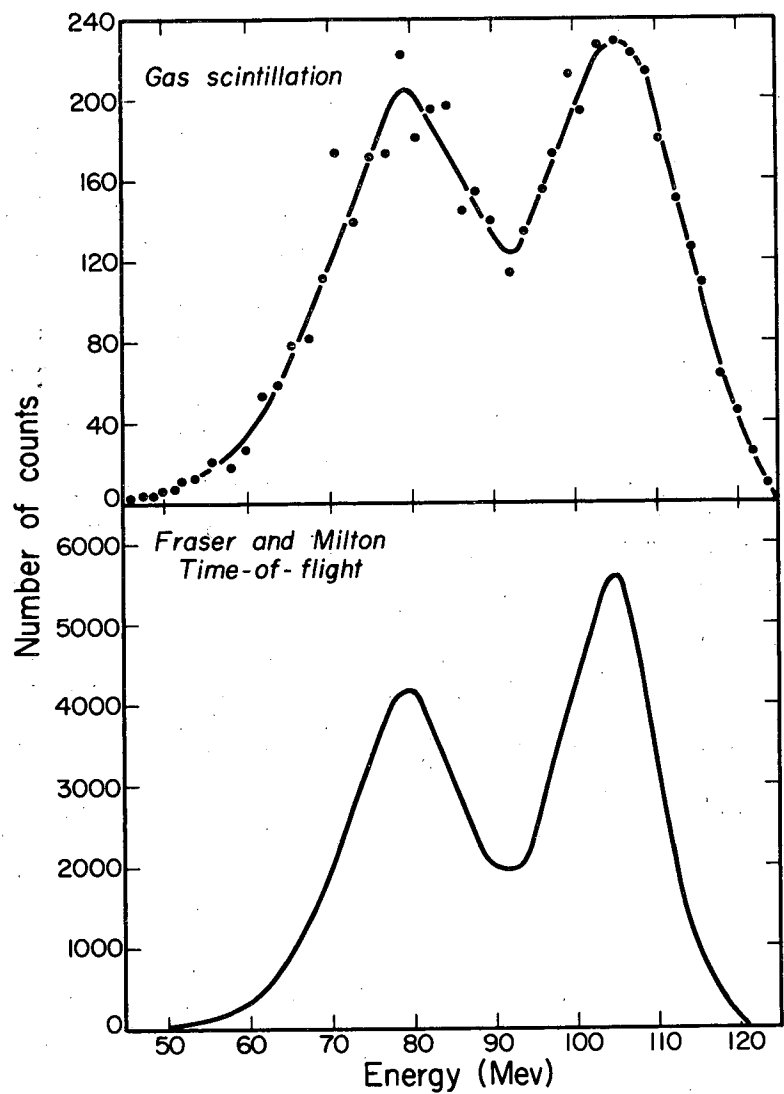
The pulse generator was included in the system in order to determine coincidence losses and to detect changes in gain and resolution due to low-energy pulses and noise in the electronic system. The pulse was triggered by a pulse from the Hilac electronic system which preceded the start of the beam burst by 2.5 msec. A variable delay in the pulse generator made it possible to produce the pulse either during the 2-msec bursts or in the interval between them. By comparing the known number of pulses sent into the system with the number that appeared under the pulse-generator peak in the spectrum from the Penco, the correction factor for coincidence loss could be determined. This factor seldom exceeded 1.2. Corrections for gain and resolution changes were determined by comparing the position and half-width of the pulse generator peak on beam-induced spectra to those of the spontaneous-fission calibration spectra.

As an example of the type of spectra obtainable with this system, the upper curve of Fig. 3 shows a fragment kinetic energy spectrum obtained



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Fig. 2. Electronic system used with the gas scintillation counter.

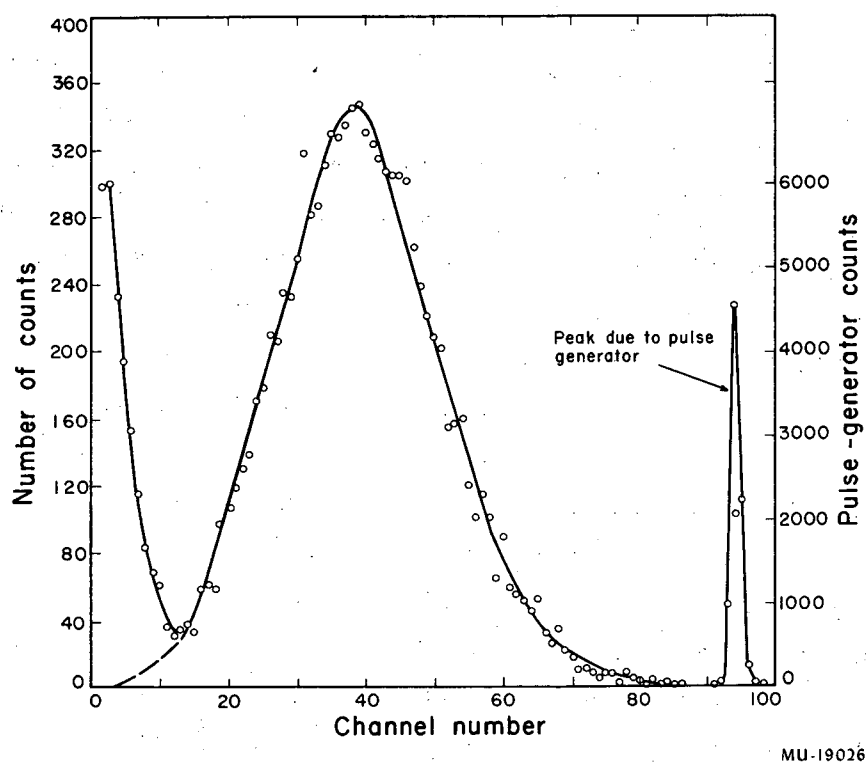


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Fig. 3. Cf^{252} spontaneous fission fragment kinetic energy spectra. Upper curve: This work, using gas scintillation counter with the sample in the window position. Lower curve: Fraser and Milton, by time-of-flight method.

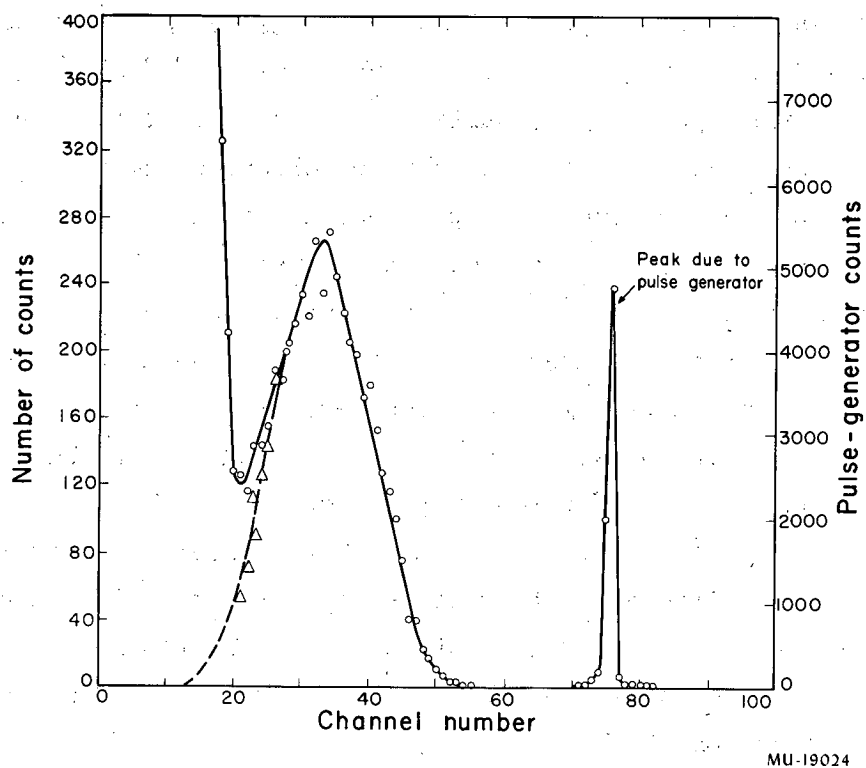
with a Cf^{252} spontaneous-fission sample in the window position of the chamber. For comparison, the lower curve of Fig. 3 shows the Cf^{252} spectrum determined by Fraser and Milton, who used a time-of-flight method.⁴³ Because of the nature of the fission-fragment kinetic energy distribution, it is difficult to assign a value for the resolution of the counter, but a reasonable estimate is 15%. This was rather high, but quite adequate for these experiments. It should be noted that if a curve for energy vs. pulse height is constructed by using the two peak positions as calibration points, the curves obtained with the sample in the window position do not extrapolate to the origin; rather, zero pulse height corresponds to approx. 8 to 10 Mev. This is apparently due to loss of light in the depression around the window, for when the sample is inside the chamber, then within the limits of experimental error one obtains a linear dependence of pulse height on energy, with zero intercept.

Examples of beam-induced fission-fragment spectra are shown in Figs. 4 and 5. At high bombarding energies, the large number of counts in the low-energy region resulted from "pile-up" (coincidence of two or more pulses in the electronic system) of low-energy pulses. This was determined by reducing the beam level, which resulted in a decrease in the ratio of low-energy pulses to fragment counts. Since the number of pile-up pulses decreases exponentially with increasing energy, it was possible to subtract them from the spectra by plotting the curves on semilogarithmic paper and extrapolating into the fragment-pulse region. Occasionally, at low bombarding energies, where the beam particles lose more energy per unit path length, the elastic-scattering peak appeared in the low-energy tail of the fragment spectra.



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Fig. 4. Fragments from fission of Au^{197} with 123-Mev C^{12} ions. Observed at 90 deg to the beam.



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Fig. 5. Fragments from fission of Au^{197} with $164.5\text{-Mev } \text{O}^{16}$ ions. Observed at 30 deg to the beam. Triangular points were obtained by logarithmic subtraction of the pile-up pulses.

2. Solid-State Conduction Counters.

a. Background

In recent years several attempts have been made to use non- or semiconducting crystals as radiation detectors. When a charged particle passes through such a crystal it loses energy by raising electrons to the conduction band of the crystal, leaving a positive "hole" in the spot vacated by the electron. By suitable application of voltage across the crystal, the electrons and holes may be collected and the resulting pulse electronically counted or pulse-height analyzed. In theory, it should be possible to obtain much better energy resolution from a solid-state detector than from a gas-filled ionization chamber. This is because much less energy is expended to form an electron-hole pair in a crystal than to form an ion pair in a gas counter. For example, it requires about 30 ev for ion-pair formation in most gases used in ion chambers, whereas, in a silicon crystal, it takes only approx 3 ev for each electron-hole pair formed. Thus, for the same amount of energy deposited in each type of detector, a silicon crystal yields about ten times as much charge. Therefore, on the basis of statistics alone, the resolution of the solid-state counter should be $\sqrt{10}$, or roughly 3 times as good as that obtained with an ionization chamber.

Much of the early work in the field of solid-state detectors is described in review articles by Champion,⁴⁴ Hofstadter,⁴⁵ and Chynoweth.⁴⁶ The counting properties of crystals such as diamond, AgCl, AgBr, CdS, and NaCl were studied. In these experiments, electric fields of the order of 5000 v/cm were generally used to collect the charges. None of the substances proved very satisfactory, as several difficulties were encountered. For one thing, the holes are often not very mobile, and thus are collected slowly if at all. Also, there are many imperfections in the crystals which may trap the electrons or holes or both before they reach the edge of the crystal. The trapped electrons and holes eventually polarize the crystal so that subsequent pulses are smaller. Various methods have been used for depolarization of the crystals, but these are rather inconvenient.⁴⁷⁻⁴⁹

The studies in which semiconductor crystals containing internal potential barriers were used have been much more successful. The first reported work is that of McKay, who used germanium containing an n-p barrier to detect alpha particles.⁵⁰ Several authors have reported the study of gold-doped germanium crystal detectors and germanium p-n junctions.⁵¹⁻⁵⁶ In general, it has been found that the signal-to-noise ratio is a sensitive function of the temperature. Thus, while the detectors yield very good energy resolution at low temperatures, noise becomes a problem at room temperature. Larger signal-to-noise ratios and better resolution at room temperature have been obtained by use of gold-doped silicon crystals. This is to be expected on the basis of the larger energy gap in the electron energy levels in silicon (approx 1.1 ev, compared with 0.72 ev for germanium).⁵⁷

Several authors have measured the amount of energy lost by ionizing particles per electron-hole pair formed.^{50,53,54,58-61} For silicon and germanium these reported values range from 2.5 to 6 ev, with the lower numbers seeming more accurate.

b. P-n Junctions

In order to discuss p-n junction detectors, it is first necessary to describe some of the properties of p-n junctions more generally. It is not possible to do this in great detail, so the reader is referred to other works on this subject.^{51,62,63} The basic material of p-n junction detectors is normally a Group IV element, and in particular, silicon or germanium. When a small concentration of a Group III element, such as boron, is added to a silicon crystal, there is an electron missing on each of the trivalent boron atoms. That is, the silicon atoms are surrounded by eight (shared) valence electrons, whereas the boron atoms have only seven.

In pure silicon it requires 1.1 ev to raise an electron from the valence band to the conduction band of the crystal. Both the electron and the positive hole left by it in the lattice may act as current carriers. If boron is present, only about 0.05 ev is required to raise an electron from the silicon valence band and place it on the boron atom.⁶⁴

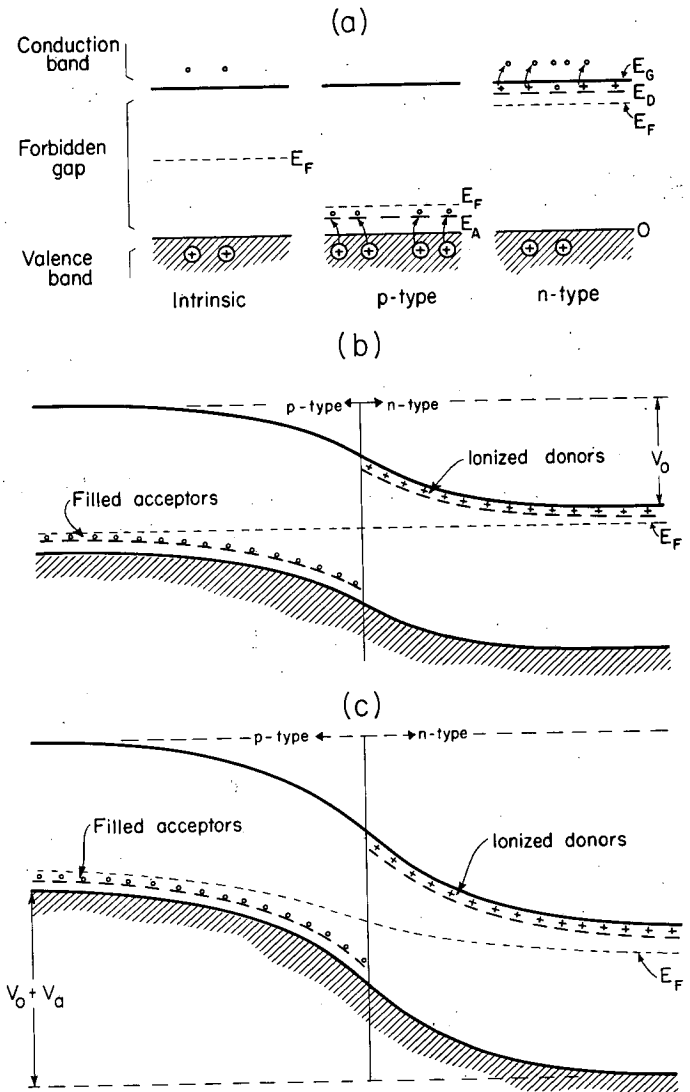
The positive hole left behind is a current carrier, but the electron on the boron is stationary. Because of their ability to accept electrons, Group III impurities are called "acceptors." A silicon crystal containing acceptor impurities is designated as "p-type," because the conduction is by positive holes.

Similarly, Group V elements, such as phosphorus, have one extra valence electron per atom. Only about 0.04 ev is needed to raise the electron into the conduction band.⁶⁵ This type of impurity is known as a "donor," and a silicon crystal doped with such impurity is called "n-type," because the conduction is by electrons.

In order to describe the effects that arise when p- and n-type crystals are joined, it is necessary to refer to the energy-level diagrams that represent them. These are shown in Fig. 6a.

In intrinsic silicon, the Fermi energy, E_F , is one-half the energy gap, E_G , above the top of the valence band. At room temperature, the acceptors in p-type silicon are nearly filled and a few electrons are present in the conduction band. As a result, the Fermi level is generally slightly above E_A , the height of the acceptor levels above the top of the valence band. The concentration of electrons in various levels and the position of the Fermi level depend upon the concentration of the impurities. Similarly, at room temperature, most of the electrons from the donor atoms in n-type silicon are excited to the conduction band, and a few have been raised from the valence band. Thus, the Fermi level is generally slightly below E_D , the height of the donor levels above the top of the valence band.

If p- and n-type silicon crystals are brought together to form a p-n junction, the electrons from the conduction band of the n-type material flow into the p-type material and fill up any empty acceptor levels and most of the positive holes in the valence band in the region near the junction. This causes p-type material to become negatively charged and the n-type positively charged. Thus, the electron energy levels are raised on the p side of the junction and lowered on the n side. When equilibrium is reached, the Fermi level is at the same height on both sides of the junction. This is shown in Fig. 6b.



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Fig. 6. Schematic electron energy level diagrams.
 (a) Intrinsic, p-type, and n-type semiconductors.
 (b) p-n junction, no applied potential.
 (c) p-n junction, reverse-biased.

The potential difference between the two sides of the junction, V_o , is given by

$$V_o = \frac{E_{Fn} - E_{Fp}}{e}, \quad (\text{II-1})$$

where E_{Fn} = Fermi energy in n-type material before forming the junction,

E_{Fp} = Fermi energy in p-type material before forming the junction,

and e = charge of the electron.

Although the potential difference is quite small (approx 0.7 v in silicon), the change takes place over a very small distance so that the resulting electric fields may be as high as 10^3 to 10^4 v/cm.

The use of p-n junctions as radiation detectors depends on the fact that there are very few current carriers in the region of the junction. This region of high resistivity is called the "depletion layer" because of its lack of mobile charges.

If the junction is reverse-biased, that is, if positive potential is applied to the n-type material and vice versa, the electron energy levels on the p-type side are raised relative to those of the n-type side. This is shown in Fig. 6c. This potential causes the current carriers to be removed at greater distances on each side of the junction, thus increasing the width of the depletion layer, or barrier. The width of the barrier, W , is given by

$$W = 1.05 \times 10^{-6} \left[\frac{\epsilon(V_o + V_a)}{\pi e N} \right]^{1/2} \text{ cm}, \quad (\text{II-2})$$

where ϵ = dielectric constant of the semiconductor,

V_o = the contact potential barrier in the absence of applied voltage,

V_a = applied reverse-bias,

and N = concentration of impurities in the silicon.

This equation is based on the assumptions that (a) there is no n-type impurity in the p-type material, and vice versa; (b) the change from

p-type to n-type material occurs in a distance short compared with the barrier width; and (c) the junction is symmetrical, that is, there are equal concentrations of impurities on the two sides.

In practice, however, p-n junctions are not normally constructed in accord with the three assumptions listed in the preceding paragraph. Assumption (a) is never absolutely obeyed, as it is not possible to completely remove all the p- or n-type impurities. However, the concentration of the major impurity is generally so large compared with that of the other type that one can ignore the minor-impurity concentration. Assumption (b) is often invalid because of the method by which the junction is constructed. It is not possible to take two semiconductors of opposite type and physically join them to form a p-n junction that has a continuous lattice structure. The junctions are often constructed by starting with a semiconductor of one type and introducing a high concentration of the opposite type of impurity into the lattice on one side of the crystal. The resulting junction is thus "smeared out" over some finite distance. For such junctions the barrier width, at low voltages, may be proportional to $(V + V_a)^{1/3}$, becoming proportional to $(V_o + V_a)^{1/2}$, in agreement with Eq. (II-2), at higher voltages.

In many instances, it is neither possible nor desirable to have the junction obey the third assumption. The junctions are often produced by diffusion of one type of impurity into a crystal containing a uniform concentration of the opposite type of impurity. In such crystals, the concentration of the diffused impurity, C_x , as a function of distance x from the face of the crystal is given by

$$C_x = C_o \left[1 - \frac{1}{\sqrt{\pi Dt}} \int_0^{x/\sqrt{Dt}} \exp(-x^2/4Dt) dx \right], \quad (\text{II-3})$$

where C_o = concentration at the surface,

t = time of diffusion process,

and D = diffusion constant of the impurity.

When the applied reverse bias on an unsymmetrical junction is increased, the depletion layer is extended into the two sides by magnitudes inversely proportional to the impurity concentration in each given material. For junctions in which the ratio of the impurity concentrations is quite large, the increase in depletion-layer width is mainly in the direction of the material having a low concentration of impurities. For these junctions, the barrier width is given by

$$W = 1.05 \times 10^{-6} \left[\frac{\epsilon(V_o + V_a)}{2\pi eN} \right]^{1/2} \text{ cm}, \quad (\text{II-4})$$

where N = impurity concentration (atoms/cm³) on the low-impurity side.

This equation may be expressed in more operational terms as

$$W = 1.44 \times 10^{-6} (\rho\mu)^{1/2} (V_o + V_a)^{1/2} \text{ cm}, \quad (\text{II-5})$$

where ρ = resistivity of the low-impurity-concentration material in Ω cm,

μ = the sum of the mobilities of the charge carriers in the lattice.

The depletion layer, being devoid of mobile current carriers, acts as a dielectric medium between the plates of a capacitor. Because of the dependence of the depletion layer upon voltage, the capacitance C of a p-n junction is dependent upon the applied reverse bias, and is given by

$$C = \frac{A\epsilon}{4\pi W}, \quad (\text{II-6})$$

where A = cross-sectional area of the junction.

c. P-n Junctions as Radiation Detectors

Because of the absence of mobile current carriers in the depletion layer, a reverse-biased p-n junction offers very high resistance to the flow of current. However, when ionizing radiation passes through this layer, it may lose energy by raising electrons from the valence band to the conduction band of the crystal; this process is similar to the production of ion pairs in a gas. Under the influence of the junction and applied potentials (as shown in Fig. 6c), the resulting electrons and

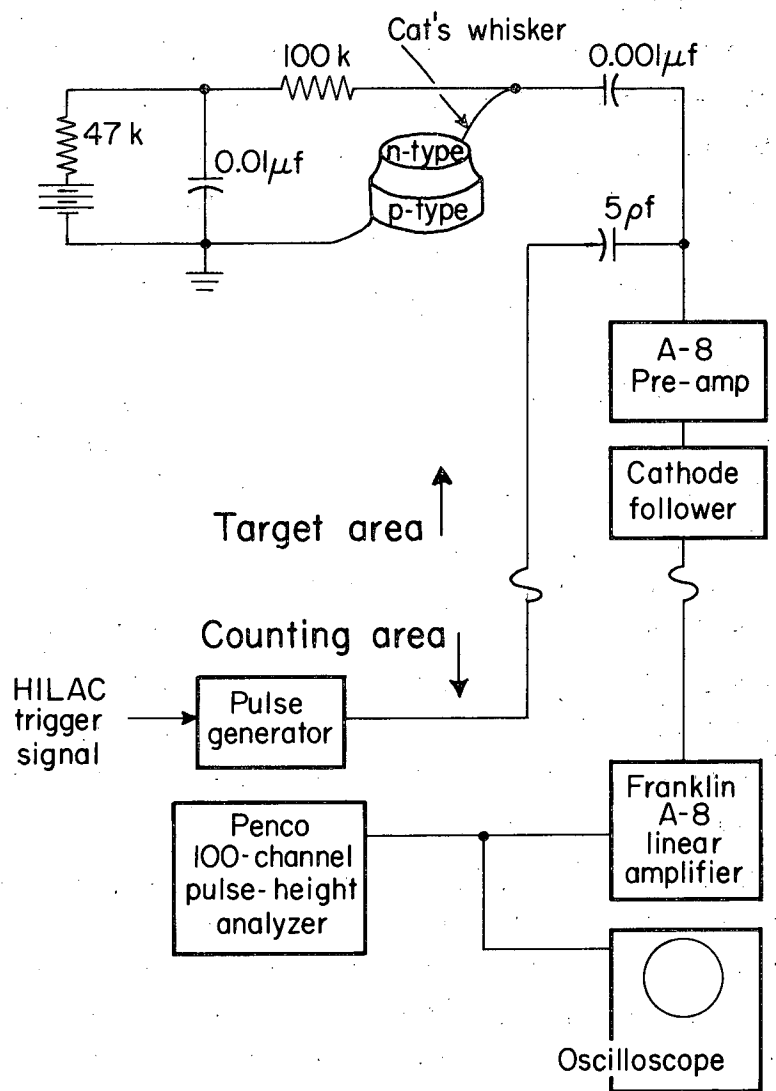
positive holes travel in opposite directions through the crystal, giving a voltage pulse which may be used to trigger a counter or may be recorded in a pulse-height analyzer.

P-n junctions have been used much more successfully than other types of solid-state counters for a number of reasons. For one thing, it is possible to maintain a very high electric field in the depletion layer with very low current flow. The positive holes in silicon semiconductors are quite mobile, their mobility being from a quarter to one-half that of the electrons. This fact, coupled with the presence of more perfect lattice structures, has eliminated the problem of polarization, encountered in previous investigations with other types of solid-state counters. Also, p-n junctions have an advantage over all other types of detectors in that their pulse rise times are quite short. The rise time, governed by the time required by the current carriers to cross the depletion layer, is dependent upon the applied voltage and the temperature, and may be as low as 10^{-11} sec. This is a calculated value, as it is not possible to measure it experimentally with present-day techniques. Potentially, semiconductors offer better energy resolution of pulses than gas-filled ionization chambers because of the lower energy required to produce a pair of charges in the semiconductor.

d. Construction and Characteristics of the Solid-State Detectors

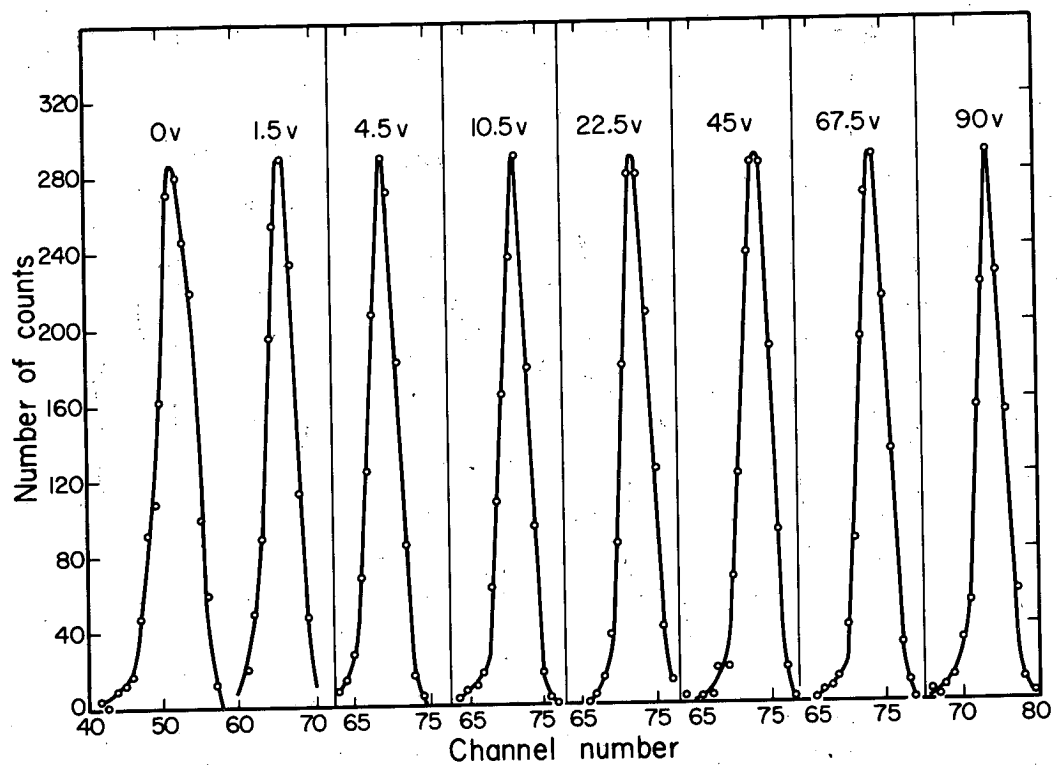
Three silicon p-n junctions were studied. Their properties are listed in Table I. The first two were produced by William Hansen (Lawrence Radiation Laboratory) and the third was obtained from Hughes Aircraft Corp. In each case the p-n junction was formed by diffusion of a high concentration of one type of impurity into one face of a crystal containing a low concentration of the opposite type of impurity. The electronic system used with these detectors is shown in Fig. 7.

Examples of the spectra obtained with these detectors are shown in Figs. 8, 9, and 10. The spectra of Fig. 8 were obtained by using a sample of Cf^{252} as a source of 6.11-Mev alpha particles. Figure 9 shows the spectra obtained at various applied voltages when 121.3-Mev C^{12} ions were elastically scattered from a thin gold target and observed at 30 deg to the beam. The energy of the carbon particles was varied by placing



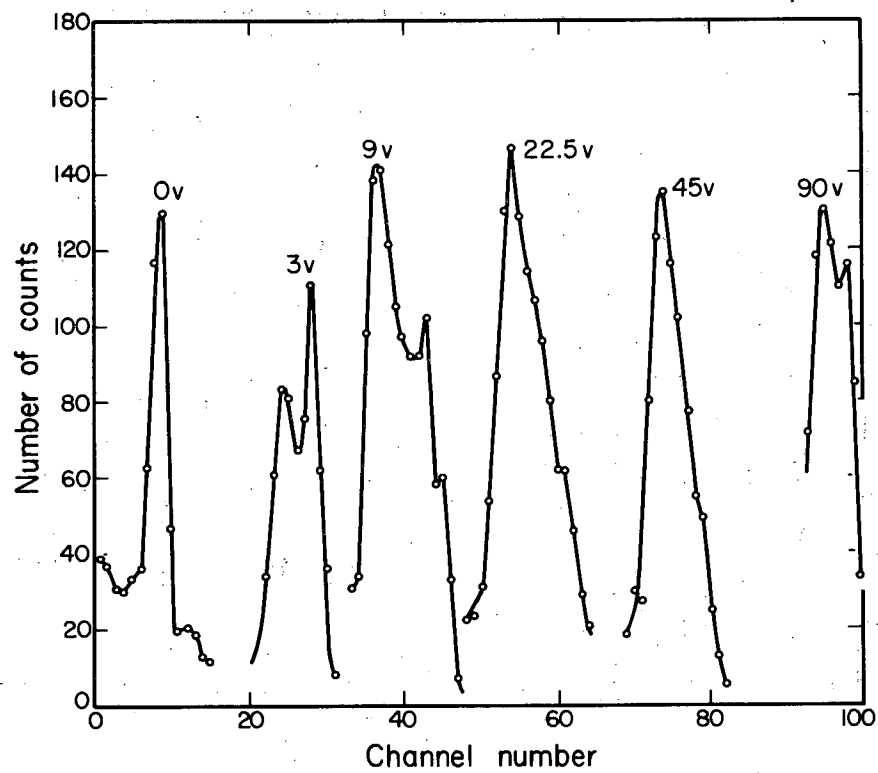
MU - 20123

Fig. 7. Electronic system used with p-n junction detectors.



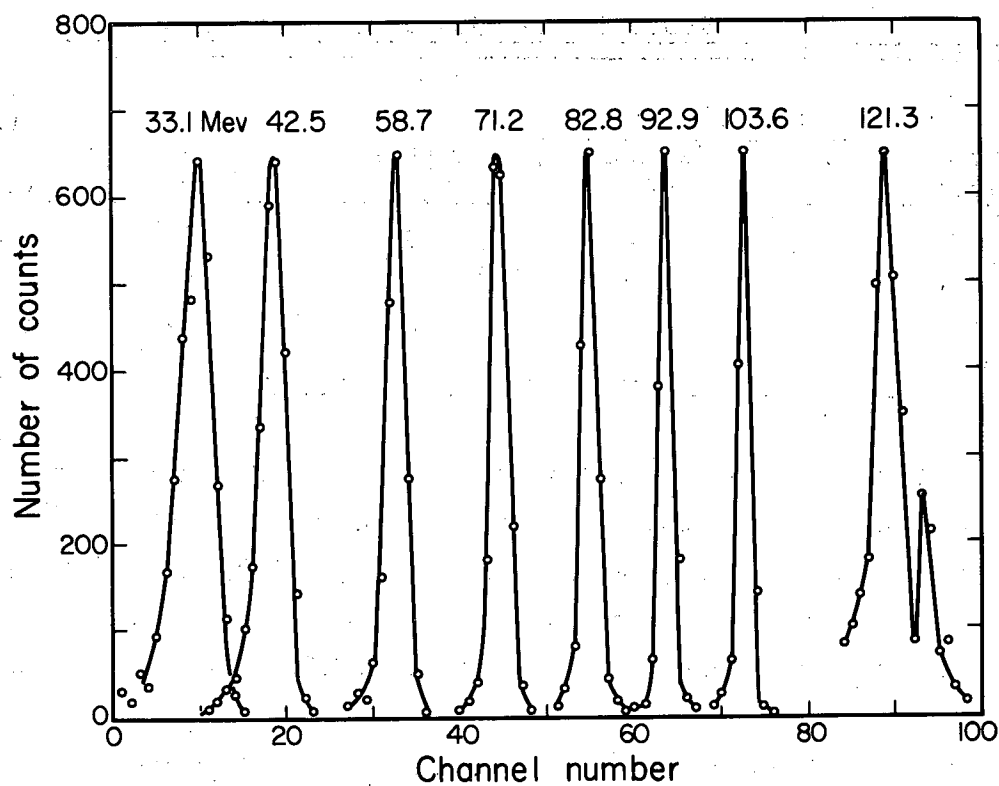
MU-20120

Fig. 8. Cf^{252} alpha-particle spectra obtained with Hu-18 p-n junction with various applied potentials.



MU - 20118

Fig. 9. Spectra observed when 121.3-Mev C^{12} particles strike the Hu-18 p-n junction with various applied potentials.



MU-20121

Fig. 10. Spectra observed when C^{12} particles of various energies strike the Hu-18 p-n junction; reverse bias is 94 v.

aluminum absorber foils in the beam path. The resulting curves are shown in Fig. 10.

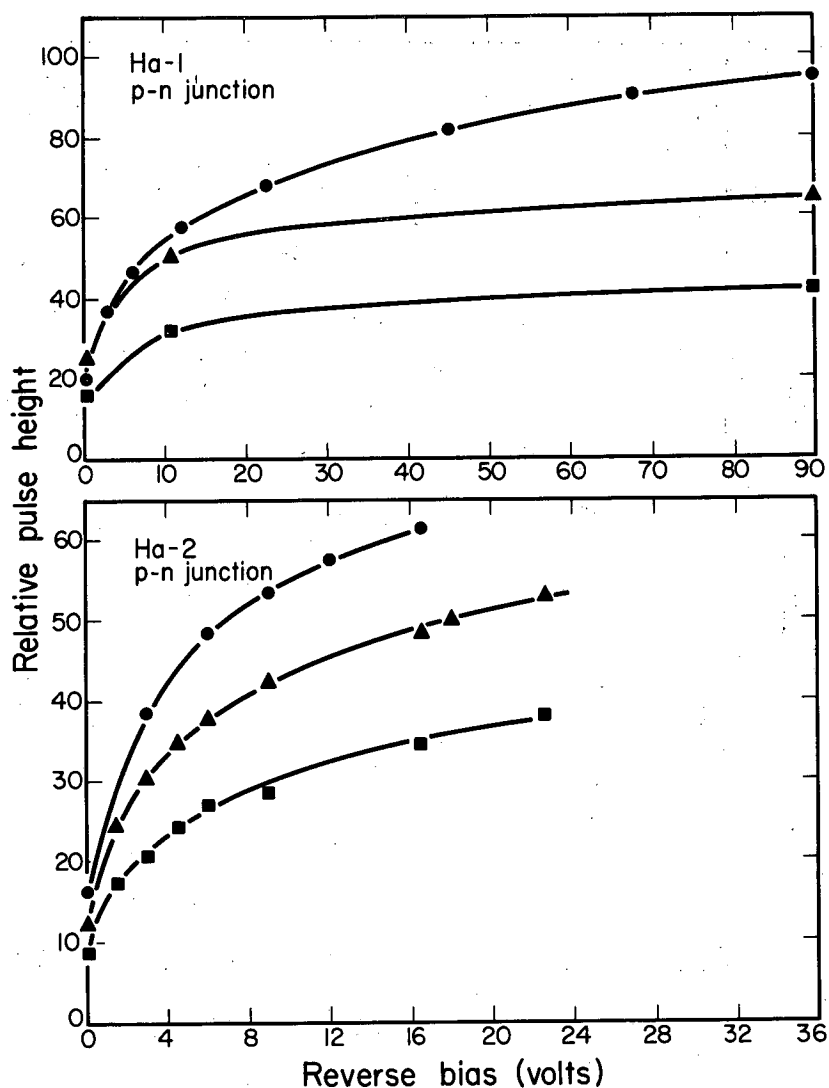
Table I. p-n junction properties

Crystal number	Base material		Diffusant type	Area (cm ²)	Est. depth of junction below surface (μ)
	Type	Resistivity (Ω cm)			
Há-1	n	100	p	~0.12	3.1
Há-2	n	100	p	~0.22	1.9
Hü-18	p	1000	n	~0.02	1.4

Admittedly the resolution of the peaks of the various spectra is not as good as that obtained in other work with semiconductor detectors.^{53,59} This is mainly because the electronic system was designed to handle the large pulses from the photomultiplier tube of the gas scintillation counter. In order to obtain better resolution it would have been necessary to lower the noise level in the electronic system and to amplify the pulses from the detector at an earlier point, preferably with a preamplifier inside the vacuum tank.

The spectra of Fig. 9 are interesting in that there appear to be two main groups of pulses. As the applied voltage is decreased, the number of counts in the higher-energy group increases relative to that of the lower-energy group. This was the only case in which such a phenomenon was observed with any of the detectors. It was known from experiments with another detector on this particular occasion that the C¹² beam contained only one energy group. This is also shown by the spectra of Fig. 10 obtained with degraded carbon beams. Other experimenters have found such effects when using a crystal which was scratched on the front surface during the polishing process.⁶⁶ Apparently it is due to some physical discontinuity in the crystal.

The variations of pulse height due to the various radiations are plotted as functions of reverse bias in Figs. 11 and 12.



MU-20117

Fig. 11. Relative pulse heights produced by alpha particles and spontaneous fission fragments as a function of reverse bias for Ha-1 and Ha-2 p-n junctions.

- - Cf^{252} alpha particles, amplifier gain approx 16.
- ▲ - Light mass group of Cf^{252} spontaneous fission fragments, gain approx 1.
- - Heavy mass group of Cf^{252} spontaneous fission fragments, gain approx 1.

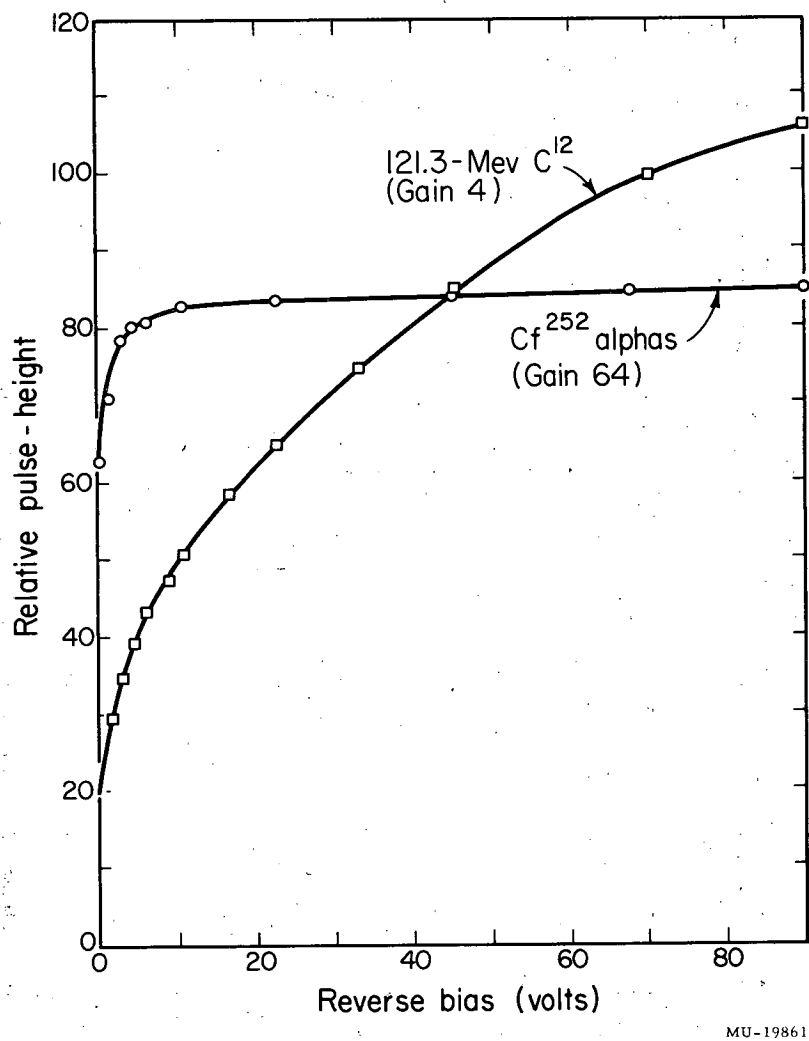


Fig. 12. Relative pulse heights produced by 121.3-Mev C¹² particles and Cf²⁵² alpha particles vs. reverse bias for Hu-18 p-n junction.

For a particle of given energy, whose range in the semiconductor material is less than the thickness of the depletion layer, the height of the pulse produced, V_{PH} , is given by

$$V_{PH} = Q/C, \quad (II-7)$$

where C is the capacity and Q is the total charge collected. The capacity is the sum of the capacity of the detector, C_d , plus the capacity of the external circuit, C_{ex} . As indicated in Eq. (II-6), C_d is inversely proportional to the depletion-layer width. The width of the depletion layer is given by Eq. (II-4) or (II-5). Collecting these terms, one expects the variation of pulse-height with applied potential to be

$$V_{PH} = \frac{Q}{C_{ex} + \frac{\text{const.}}{(V_a + V_o)^{1/2}}} \quad (II-8)$$

As noted above, the barrier width may be proportional to $(V_o + V_a)^{1/3}$ at low voltages.

As the applied voltage is increased, C_d becomes small compared with the external capacity, and the pulse height for a given number of electron-hole pairs created approaches a maximum value of Q/C_{ex} . This is clearly shown by the pulse heights produced by 6.11-Mev Cf^{252} alpha particles when observed with the Hu-18 detector (Fig. 12). The capacities of Ha-1 and Ha-2 are larger relative to C_{ex} than that of Hu-18, thus the alpha-particle pulse-height curves for those detectors have not reached their maximum values at the highest voltages studied (Fig. 11).

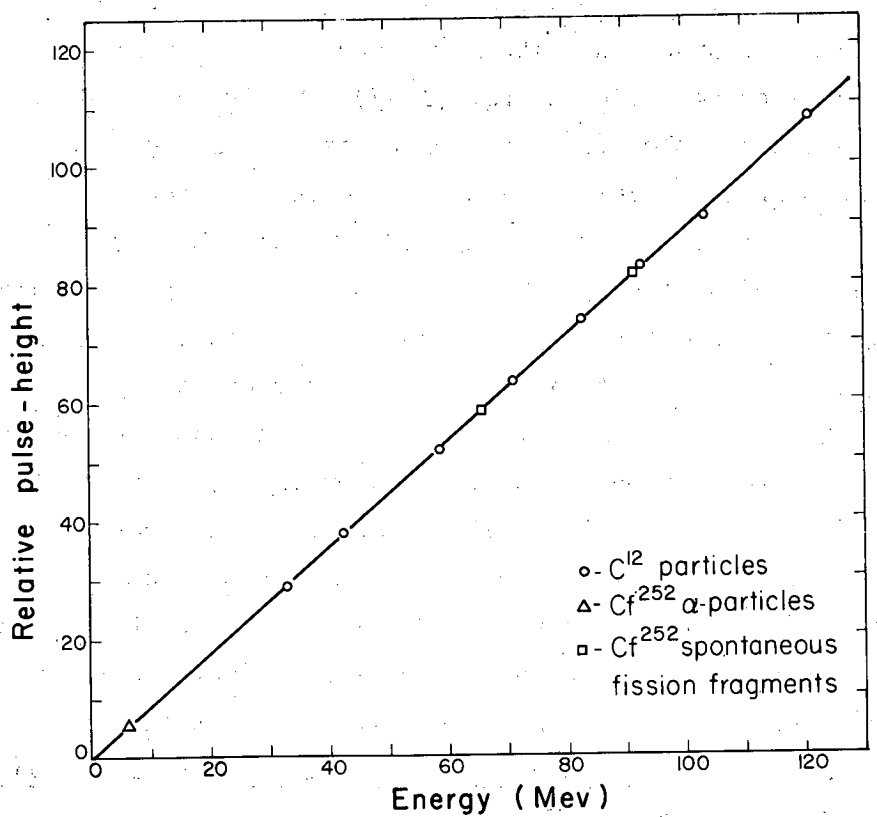
If the range of the particle being observed is longer than the thickness of the sensitive counting region, W_c , the pulse-height-vs-voltage curve rises faster than predicted by Eq. (II-8). This is because the pulse-height increase is due to an increase in W_c , thus an increase in the amount of energy deposited in W_c , in addition to the capacitance effect. This is apparently the reason why the 121.3-Mev C^{12} pulse-height curve continues to rise after the alpha-particle curve has leveled off in Fig. 12. This is discussed below in more detail.

In order to obtain a pulse-height-vs-energy relationship for C^{12} particles observed in the Hu-18 detector, the pulse heights of the peaks of the various curves of Fig. 10 were plotted as a function of the energy of the carbon particles. The resulting curve is shown in Fig. 13. It was necessary to add 19 channels to the channel number of each peak to correct the pulse heights for the threshold setting of the analyzer. This figure was determined by a pulse-generator calibration of the analyzer. Points for 6.11-Mev alpha particles from Cf^{252} and for the light- and heavy-mass groups of fission fragments from Cf^{252} have also been plotted on the curve. The alpha-particle point was obtained from the 90-volt point of Fig. 12, plus a pulse-generator determination of the ratio of amplifier gains at the two settings used. It appears in Fig. 13 that

- (a) pulse height is proportional to the energy deposited by the particle in the sensitive counting region,
- (b) alpha-particle and fission-fragment pulses fall on the same curve as carbon particles.

The first result is in agreement with previous studies of pulse heights vs energy,^{52,53,59,67} but this is the first demonstration that the relation is valid over such a wide range of particle energies. The alpha-particle result is also in agreement with previous work in which the pulse heights of protons, He^3 particles, and alpha particles of various energies were studied.^{53,67} Treatment of the fission-fragment data is discussed below.

If one assumes that the rise of pulse height with voltage for the alpha-particle curve of Fig. 12 is due entirely to the capacity effect, it is possible to correct the carbon points for the decrease in capacity. From the pulse heights thus obtained one can, from the pulse-height-vs-energy curve of Fig. 13, determine the energy deposited by the carbon particles in the sensitive counting region. By the use of range-energy curves for carbon particles in the detector material, it is possible to infer the thickness W_c of the sensitive counting region from the amount of energy deposited. Since range-energy data for carbon particles in silicon were not available, the curves for the neighboring element, aluminum, were used.⁶⁸ This is not expected to introduce any appreciable error.



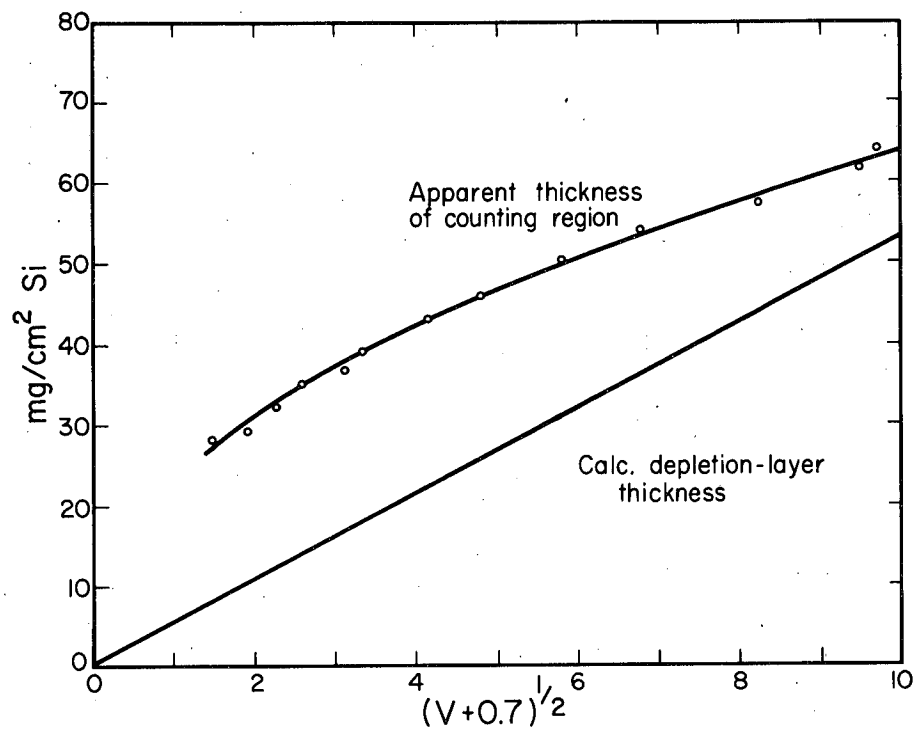
MU-19582

Fig. 13. Relative pulse height as a function of energy for C^{12} particles, and for Cf^{252} alpha particles and fission fragments. Obtained with the Hu-18 p-n junction with reverse bias of 94 volts.

The results of this analysis for the pulse heights produced by 121.3-Mev C^{12} particles, shown in Fig. 12, are shown in Fig. 14, where the apparent thickness of the sensitive region is plotted versus $(V_a + V_o)^{1/2}$, where V_o (= 0.7 volt) is the internal potential barrier of the silicon p-n junction. The lower curve is the depletion-layer thickness calculated according to Eq. (II-5). The results indicate that W_c is larger than the depletion layer thickness by a roughly constant amount. This effect arises because electrons produced in the p-type material beyond the depletion layer slowly diffuse into the barrier region and are accelerated across it. This causes a voltage pulse whose rise time is much longer than that from the charges formed in the depletion layer. However, if the pulses from the detector are not clipped with a very short time constant (RC) in the external circuit, a contribution from the electrons in the region beyond the depletion layer is included in the pulse. It is thus expected that the apparent thickness of the counting region would be decreased by use of a shorter time constant in the external circuit. This effect does not occur with the holes formed in the n-type material on the face of the detector because of rapid recombination of charges in the region of high impurity concentration.

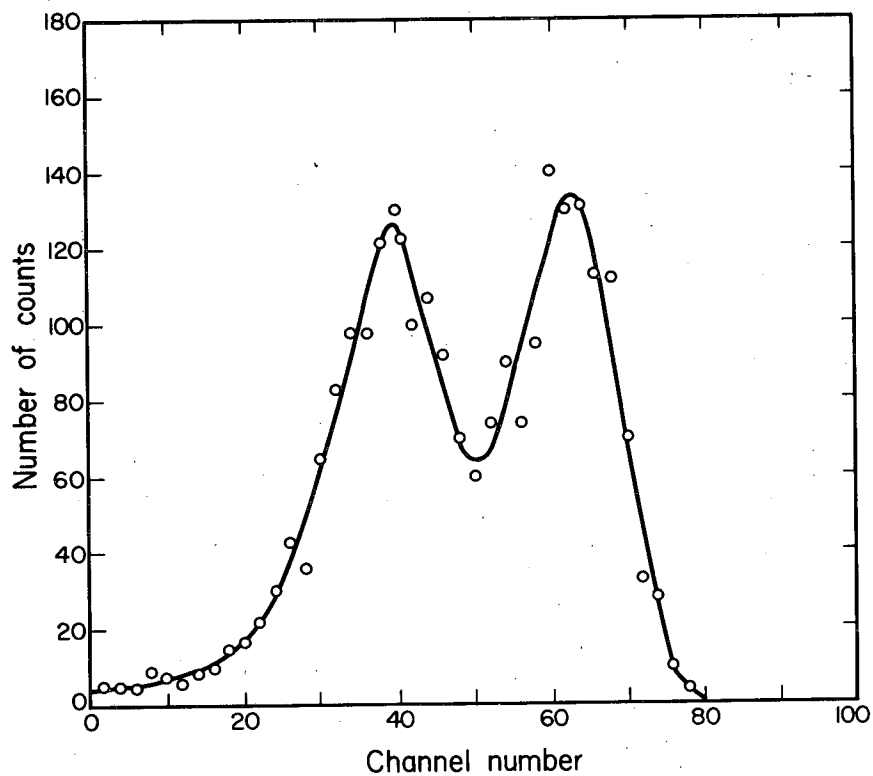
e. Use of the Solid-State Detectors as Fission-Fragment Counters

The spectra shown in Figs. 15 and 16 represent the kinetic energy distributions of fragments from spontaneous fission of Cf^{252} observed in Hu-18 and Ha-2. From Fraser and Milton's time-of-flight data it is known that the energies corresponding to the two peaks of the kinetic energy spectrum are 104.7 and 79.8 Mev.⁴³ The ratio of energies of the two groups is thus 1.31. From all the fission-fragment spectra obtained with the solid-state detectors in this study, the ratios of pulse heights are greater than 1.31. If pulse height is proportional to the amount of energy deposited by the fragments, independent of mass and atomic number, this result indicates that both fragment groups have lost energy in passing through the front surfaces of the detectors. The ratio of pulse heights of the two groups observed with Ha-1 and Ha-2 are plotted versus the applied voltage in Fig. 17. It is seen that the ratio is approximately independent of reverse bias. Thus the depletion layer increases



MU - 20125

Fig. 14. Apparent thickness of the sensitive counting region and the calculated depletion-layer thickness as a function of $(V_a + 0.7)^{1/2}$ for Hu-18 p-n junction.



MU-20134

Fig. 15. Kinetic energy spectrum of fragments from spontaneous fission of Cf^{252} observed with Hu-18 p-n junction with reverse bias of 94 v.

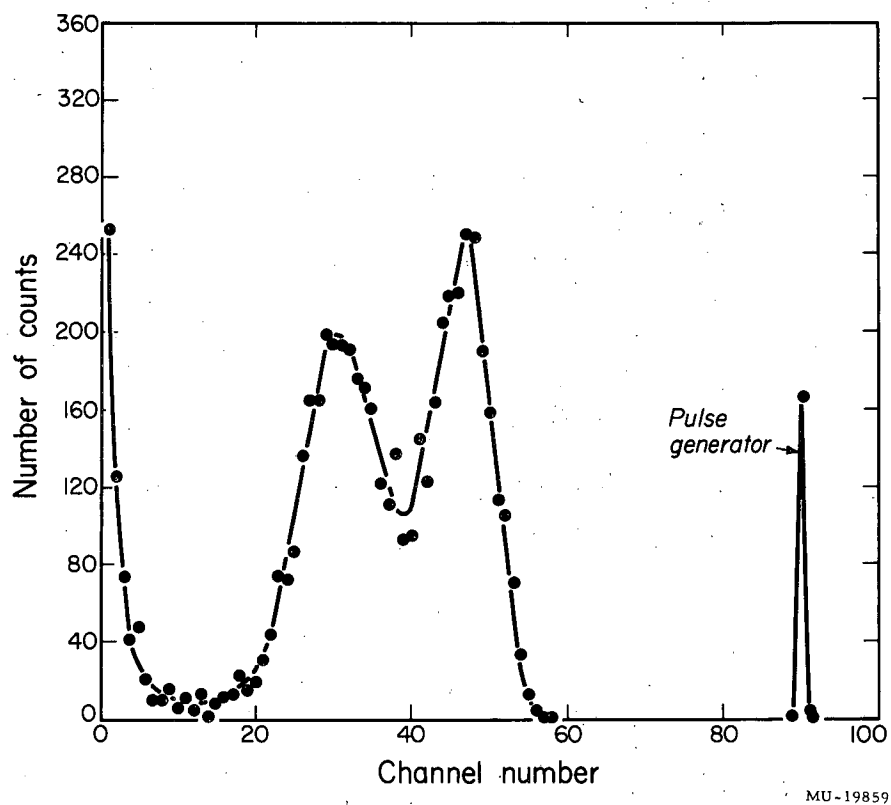
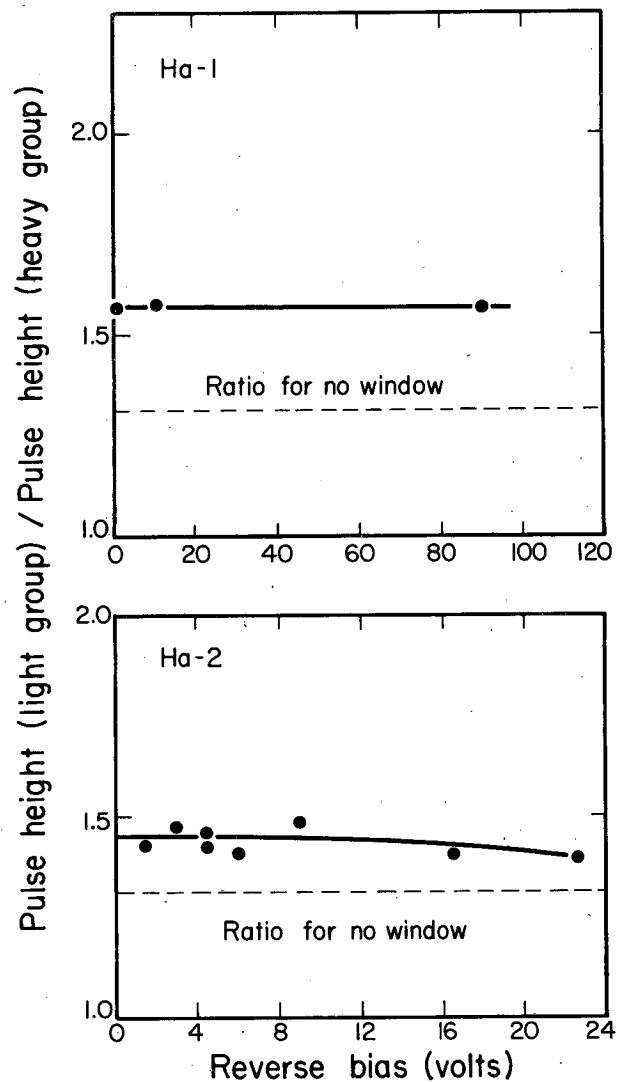


Fig. 16. Kinetic energy spectrum of fragments from spontaneous fission of Cr^{252} observed with Ha-2 p-n junction with reverse bias of 9 v.



MU - 20124

Fig. 17. Ratio of pulse heights produced by the two mass groups of Cf^{252} spontaneous-fission fragments as a function of reverse bias for Ha-1 and Ha-2 p-n junctions.

only slightly, if at all, in the direction of the front surface. This is in agreement with the ideas presented above in the discussion of p-n junction theory.

In order to determine the kinetic energy of fragments from heavy-ion-induced fission, it is necessary to make corrections for energy loss in the front surface of the detector. Two different approaches have been made to solve this problem.

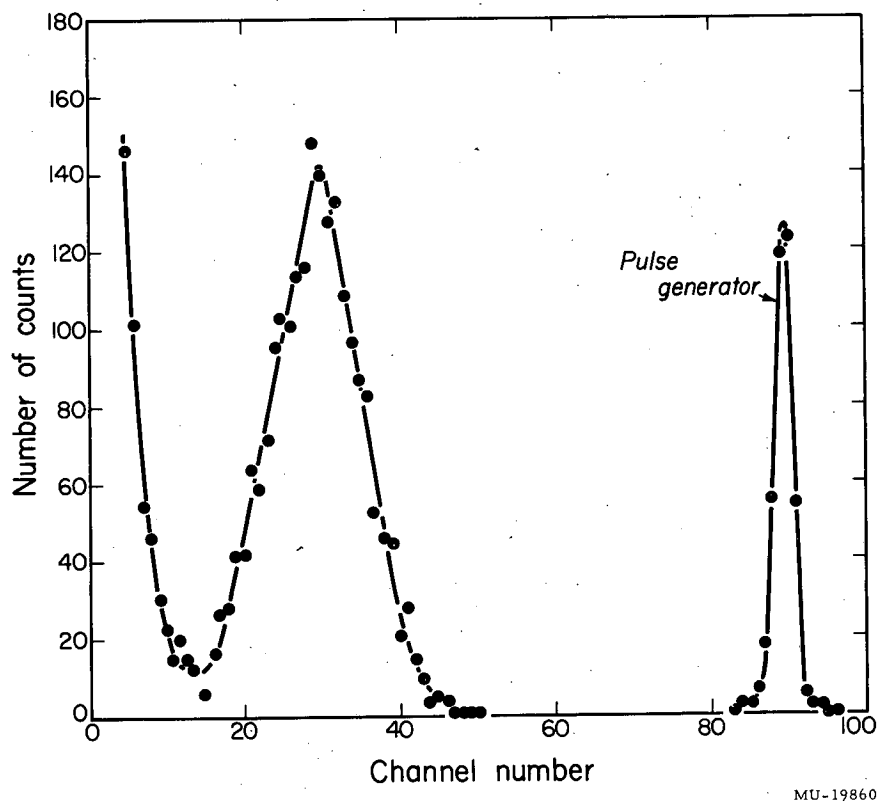
On one hand, one may assume that the pulse-height-vs-energy relationship is the same for fission fragments as for carbon particles. Thus the positions of the peaks of Fig. 15 correspond to energies of 91.4 and 65.6 Mev on the pulse-height-vs-energy curve of Fig. 13. The thickness of the "window" is then determined from the energy loss of the light-fragment group ($104.7 - 91.4 = 13.3$ Mev) and the fission-fragment range-energy data of Schmitt and Leachman.⁶⁹ The check on the self-consistency of this approach is to use the window thickness obtained for the light-fragment group to correct the energy of the heavy group. This procedure yields an energy of 80.4 Mev for the heavy group after correction. This is in good agreement with the expected value of 79.8 Mev.

The second method is to assume that the pulse-height-vs-deposited-energy curve for fission fragments is linear and passes through the origin. Various window thicknesses are assumed and energy corrections are made until the ratio of corrected energies is 1.31. The results of this analysis applied to the data of Fig. 15 are identical with those obtained using the first procedure. In each case the window thickness is found to be approx $350 \mu\text{g}/\text{cm}^2$. The producers of the crystal estimated the window thickness to be about $170 \mu\text{g}/\text{cm}^2$. In view of the method of obtaining that estimate (from surface impurity concentration, diffusion time, etc.) the discrepancy is not alarming. Another possible cause for the discrepancy would be the presence of a coating of a light element, such as oxygen, on the front surface. The estimated window thicknesses listed in Table I were obtained by this procedure.

The fission-fragment deposition energies obtained by the second method are plotted in Fig. 13. From the good agreement obtained, it

appears that energy deposited per electron-hole pair formed is the same for fission fragments as for carbon particles. If there is any "ionization defect" for fission fragments in the silicon detector it is too small to be observed by these techniques. It is expected to be small because of the small amount of energy required for electron-hole pair formation in the semiconductor material.⁷⁰

The Ha-2 detector was used for most of the heavy-ion-induced fission experiments. Of all the detectors, this one most nearly met the requirements of those experiments - physical size, resolution, low noise level, and small pulses from scattered particles. Unfortunately, all measurements with this detector were confined to voltages below 22 volts, as the noise level became severe at higher voltages. However, with 9 volts reverse bias, the fission-fragment pulses were adequately separated from scattered-particle pulses. This is shown in Fig. 18. As with the gas scintillation counter, pile-up of scattered-particle pulses in the slow electronic system was a serious problem at extreme forward angles, as shown in Fig. 19.



MU-19860

Fig. 18. Spectrum of fragment kinetic energies from fission of Au^{197} with 93-Mev C^{12} ions. Observed at 90 deg to the beam with Ha-2 p-n junction with reverse bias of 9 v.

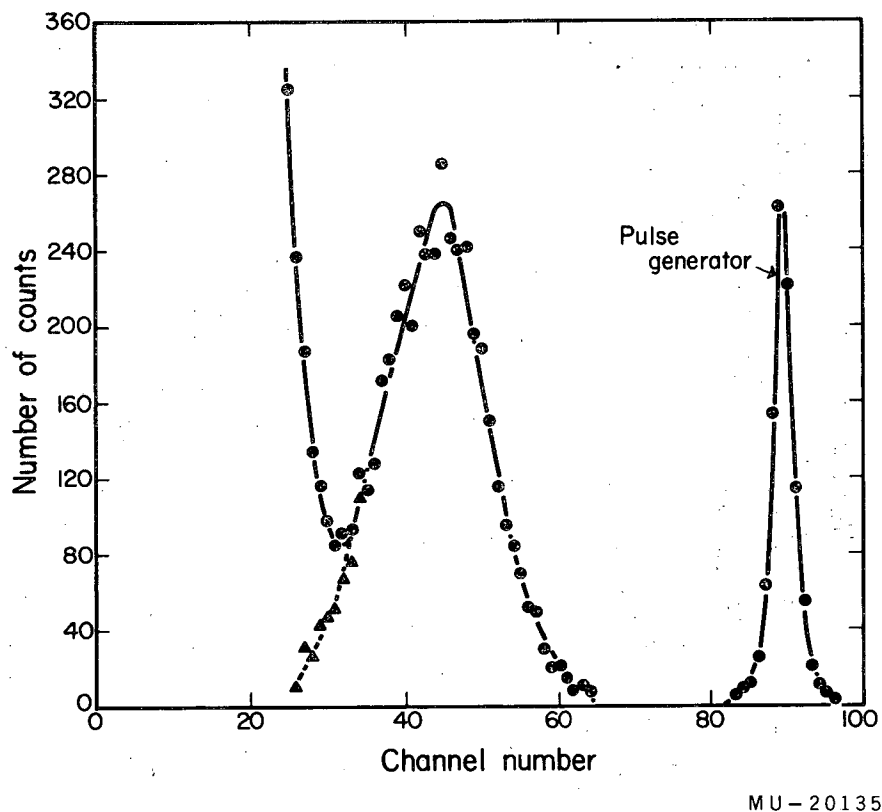


Fig. 19. Spectrum of fragment kinetic energies from fission of Au^{197} with 93-Mev C^{12} ions. Observed at 30 deg to the beam with Ha-2 p-n junction with reverse bias of 9 v. Triangular points show result of logarithmic subtraction of pile-up pulses.

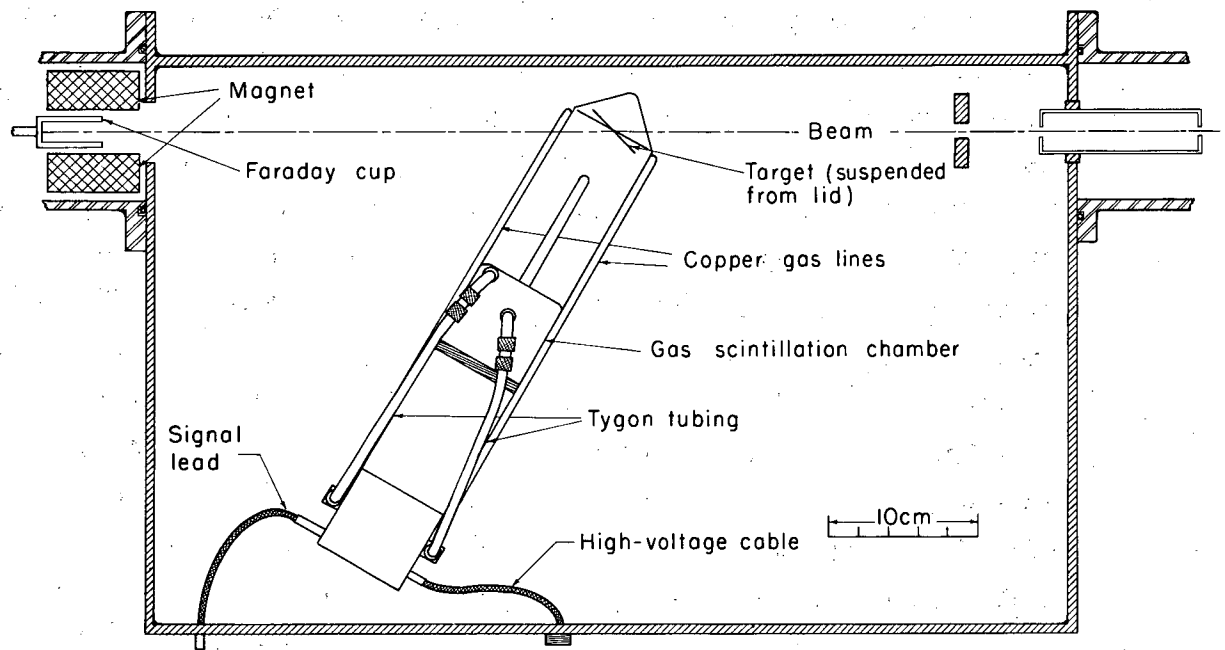
3. Bombardment Techniques

The heavy-ion beams used in these experiments were obtained from the Hilac (heavy-ion linear accelerator). This is a resonant-cavity machine which accelerates various heavy ions up to a constant velocity, regardless of mass and charge state. The emerging beam particles have energies of 10.4 Mev/nucleon, for example, 124.8-Mev C^{12} . The beam normally was bent through 15 deg by a magnetic field before being allowed to enter the target chamber. This was done in order to remove any other energy groups that might have been accelerated along with the principal energy group.

The bombardments were made in the vacuum tank shown schematically in Fig. 20. Before entering the tank, the beam passed through two 1/8-in. diameter collimators, 3 in. apart. A third collimator, 3/8 in. in diameter, inside the tank, prevented particles scattered by the second collimator from entering the counter when at forward angles.

The targets were suspended from the lid of the assembly and could be rotated from outside the tank. Targets generally consisted of 100 to 200 $\mu\text{g}/\text{cm}^2$ of fissionable material (vaporized when possible) on 0.1-mil Al foil. When fragments were counted at forward angles, the target was placed at 45 deg to the beam, with the target material facing away from the beam, and when counted at backward angles, the target faced the beam at 45 deg.

The counter was attached to a table pivoted directly under the target, which could be rotated from outside the tank without destroying the vacuum. Gas lines for the scintillation counter entered the tank through the pivot. The range of angles obtainable was mechanically limited to 23 to 156 deg for the scintillation counter, and 8 to 174 deg for the physically smaller solid-state counter. The angle subtended by the counters ($\Delta\theta$) could be varied between approx 2 and 4 deg by changing the radius. In normal use this angle was about 3 deg. In order to observe fragments at extreme forward or backward angles, the counters were backed away from the target as far as possible, thus reducing the subtended angle to approx 2 deg.



MU-19022

Fig. 20. Schematic diagram of the vacuum tank.

After passing through the target, the beam was collected in a copper Faraday cup. When absolute beam currents were desired, a magnetically shielded cup in the port at the rear of the tank was used. A discussion of the calibration and properties of Faraday cups is given in Appendix A.

For bombardments below the full energy of the Hilac beams, weighed aluminum foils were placed in the beam path ahead of the vacuum tank. The resulting energies were determined from the range-energy curves of J. R. Walton.⁶⁸

The vacuum in the tank was normally connected to that of the Hilac poststripper cavity and pressures were of the order of 5×10^{-6} mm Hg.

The "T" assembly, shown in Fig. 43 of Appendix A, was used for some of the experiments in which it was necessary to observe the fragments only at 90 deg to the beam.

B. Experimental Results and Treatment of Data

1. Types of Experiments

Two types of experiments were used to obtain the fission-fragment data. Angular distributions and kinetic energy spectra at various angles to the beam were obtained from bombardments performed in the vacuum tank. Both the gas scintillation and solid-state detectors were used in these experiments. It was feasible to obtain the complete angular distributions only at a few widely spaced bombarding energies. In order to obtain cross sections and fragment kinetic energies at intervening bombarding energies, fragments were observed only at 90 deg to the beam at many closely spaced bombarding energies. These experiments were performed in the "T" assembly, in which a gas scintillation counter was fixed at 90 deg to the beam.

2. Kinetic Energy Data

All the beam-induced fission-fragment kinetic energy spectra observed in this work were approximately symmetric in distribution. This is apparently the result of mass-yield distributions corresponding to symmetric fission in all the systems investigated. In interpreting the kinetic energy data it has been assumed that the most probable kinetic energy corresponds to the average kinetic energy per fragment for symmetric division of the fissioning nucleus. The calculations discussed in Appendix B indicate that this assumption is justified.

Both types of fission-fragment detectors were calibrated periodically during a series of experiments by taking spectra from the Cf^{252} spontaneous-fission source. A curve of pulse height vs fragment energy deposited in the gas scintillation chamber was obtained by observing the Cf^{252} spectrum with the sample in the window position. A spectrum was then obtained with the sample outside the window. The effective window thickness was determined from the energy lost by the two fragment groups in passing through the window, and from range-energy data of Fulmer for fission fragments in nickel.⁷¹ The effective window thickness determined in this manner was about 10% greater than the weighed thickness, apparently owing to buckling of the nickel foil under the 1-atmosphere pressure difference.

In this work an attempt was made to determine fragment range-energy curves in nickel by observing the positions of the peaks corresponding to the two Cf²⁵² fragment groups as a function of the thickness of nickel placed between the source and the scintillation detector. The results of these experiments indicated that dE/dx of the fragments remains nearly constant, decreasing only slightly between 100 and 30 Mev. This is in very poor agreement with Fulmer's data, which indicate a general decrease of dE/dx with fragment energy. The reason for this discrepancy is not understood. In principle, Fulmer's results should be more accurate because he used a narrow spread of fragment masses, whereas in these experiments a wide distribution of masses was always represented in the spectra. For this reason, the data of Fulmer have been used in making corrections for energy loss in the window. The determination of the p-n-junction "window" thickness with the aid of the Schmitt and Leachman data on fragment range energy in aluminum has been described in the discussion of solid-state counters.⁶⁹

Before correction of the beam-induced fragment kinetic energies for passage through the window, corrections for any gain shifts in the electronic system were made by noting the position of the pulse-generator peak. Corrections for energy loss in the target material were made with the aid of the empirical curves discussed in Appendix A.

After these corrections were made, the most probable kinetic energy was plotted as a function of laboratory-system angle. The results of this analysis for Au + C¹² at three bombarding energies, and for Au + 164.5-Mev O¹⁶, are shown in Figs. 21 and 22. The limits of error shown on the graphs represent uncertainty in determination of the channel number of the peaks of the kinetic energy spectra.

For a given energy in the center-of-mass system and value of η , the energy in the laboratory system as a function of angle is given by (cf. Appendix B)

$$E_{\text{lab}} = E_{\text{cm}}(1 + \eta^2 + 2\eta \cos\theta_{\text{cm}}). \quad (\text{II-9})$$

The quantity η is defined by the equation

$$\eta = \frac{v_{\text{comp}}}{v_{\text{cm}}}, \quad (\text{II-10})$$

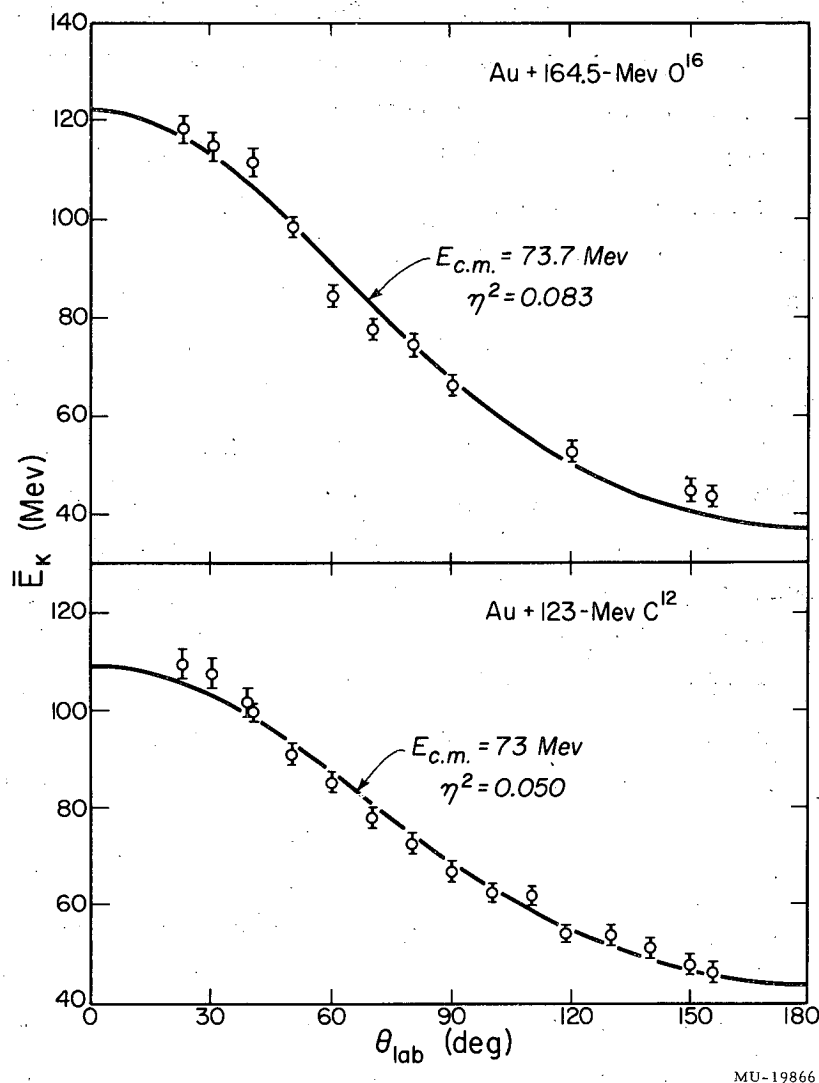


Fig. 21. Most probable kinetic energies as a function of laboratory-system angle. Upper curve: Au + 164.5 Mev O^{16} ; lower curve: Au + 123-Mev C^{12} . Experimental points from data taken with the gas scintillation chamber.

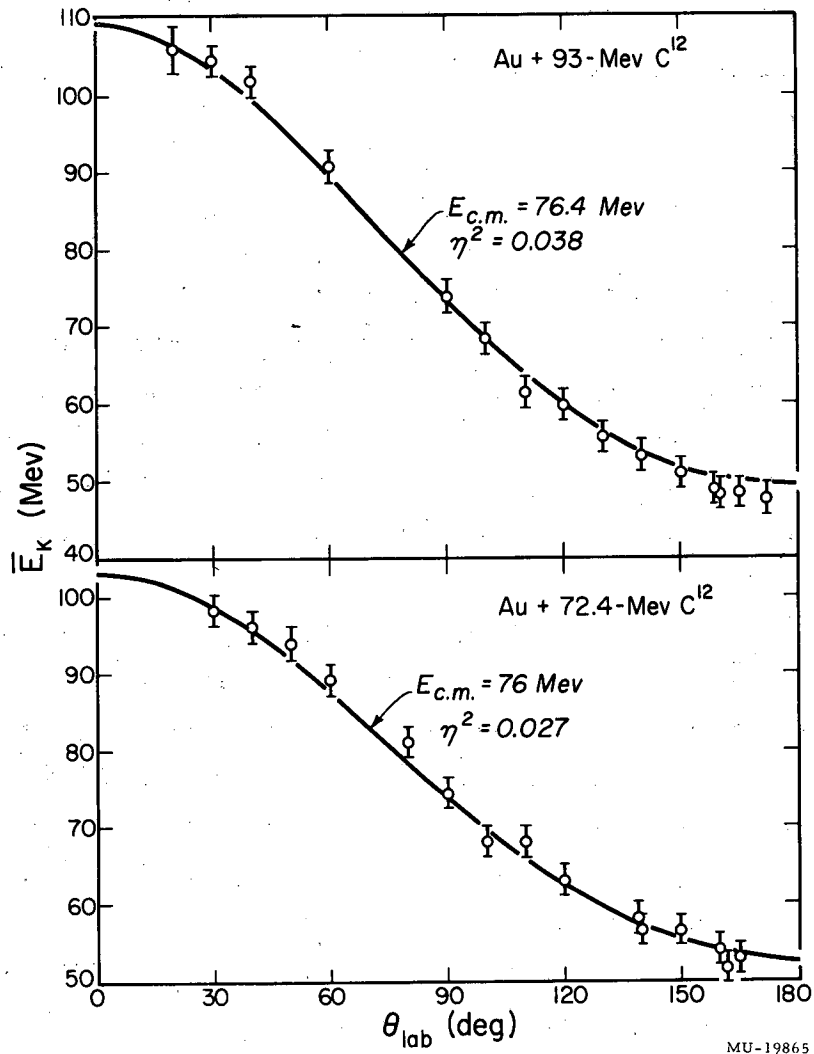


Fig. 22. Most probable kinetic energy as a function of laboratory-system angle. Upper curve: Au + 93.3-MeV C¹²; lower curve: Au + 72.4-MeV C¹². Experimental points from data taken with Ha-2 solid-state detector.

where v_{comp} = velocity of the compound nucleus in the direction of the beam,

and v_{cm} = velocity of the fragment in the center-of-mass system.

The center-of-mass and laboratory angles are related by the expression

$$\tan\theta_{\text{lab}} = \frac{\sin\theta_{\text{cm}}}{\eta + \cos\theta_{\text{cm}}} \quad (\text{II-11})$$

Various values of E_{cm} and η were used in Eq. (II-9) to find the values that give best agreement with the experimental points of Figs. 21 and 22. The resulting calculated curves are shown in the figures.

Several observations may be made concerning these results. In general, the kinetic energies obtained with the solid-state detector are higher than those obtained from the gas-scintillation data. This is probably due to the window corrections applied to the solid-state-detector data. The window thickness for that counter was estimated on the basis of assumptions which cannot be fully justified until detectors are available in which the sensitive counting region extends to the front surface of the crystal. For this reason, the energy data from the gas scintillation chamber are considered more reliable, although, as noted above, the range-energy data for nickel are subject to some question.

There is reasonable agreement between the calculated curves and the experimentally determined kinetic energies, although at the forward angles of the curves for 123-Mev C^{12} and 164.5-Mev O^{16} bombardments, the experimental points appear to be too high. This error could arise from inaccuracy of the window corrections, as the range-energy curves rise steeply in this energy region and Fulmer's data must be extrapolated for energies above 100 Mev. The discrepancy could also result from increased gain of the photomultiplier tube due to the high counting rate of scattered particles at small angles. Diamond has observed such a phenomenon with this type of photomultiplier tube.⁷²

If there is a complete transfer of momentum by the bombarding particle to the compound nucleus, η is given by the equation

$$\eta^2 = \frac{A_b E_b}{A_{\text{comp}}^2} \frac{A_{\text{frag}}}{E_{\text{cm}}}, \quad (\text{II-12})$$

where A_b and E_b are the mass and energy of the bombarding particle, A_{comp} is the mass of the compound nucleus, and A_{frag} and E_{cm} are the mass and center-of-mass kinetic energy of the fragment. Within the limits of experimental error, the η values used to fit the E_{lab} -vs- θ_{lab} data agree with the values calculated for full momentum transfer. In calculation of the values for full momentum transfer, E_{cm} has been obtained from the gas-scintillation data, and most probable fragment masses of 100 and 102 have been assumed for C^{12} - and O^{16} -induced fission of gold, respectively. These values have been estimated from the mass-yield data of Tarantin et al. for 115-Mev N^{14} -induced fission of gold.³⁷ A summary of the η values obtained by the various methods is given in Table II.

Table II. Values obtained for the quantity η

System	From E_{lab} vs θ_{lab}	From angular distribution	Full momentum transfer
Au + 164.5-Mev O^{16}	0.288 ± 0.01	0.288	0.284
Au + 123.3-Mev C^{12}	0.224 ± 0.01	0.223	0.218
Au + 93.3-Mev C^{12}	0.195 ± 0.01	0.188	0.192
Au + 72.4-Mev C^{12}	0.164 ± 0.005	0.164	0.170

In order to more carefully determine the variation of kinetic-energy release as a function of bombarding-particle energy, fragments were observed at 90 deg to the beam in the "T" assembly for the systems Au + C^{12} and Pt + N^{14} at various bombarding-particle energies. In the platinum experiments, a target enriched in Pt^{198} was used. Its composition is given in Section III.

At 90 deg to the beam, the relation between E_{cm} and E_{lab} is given by

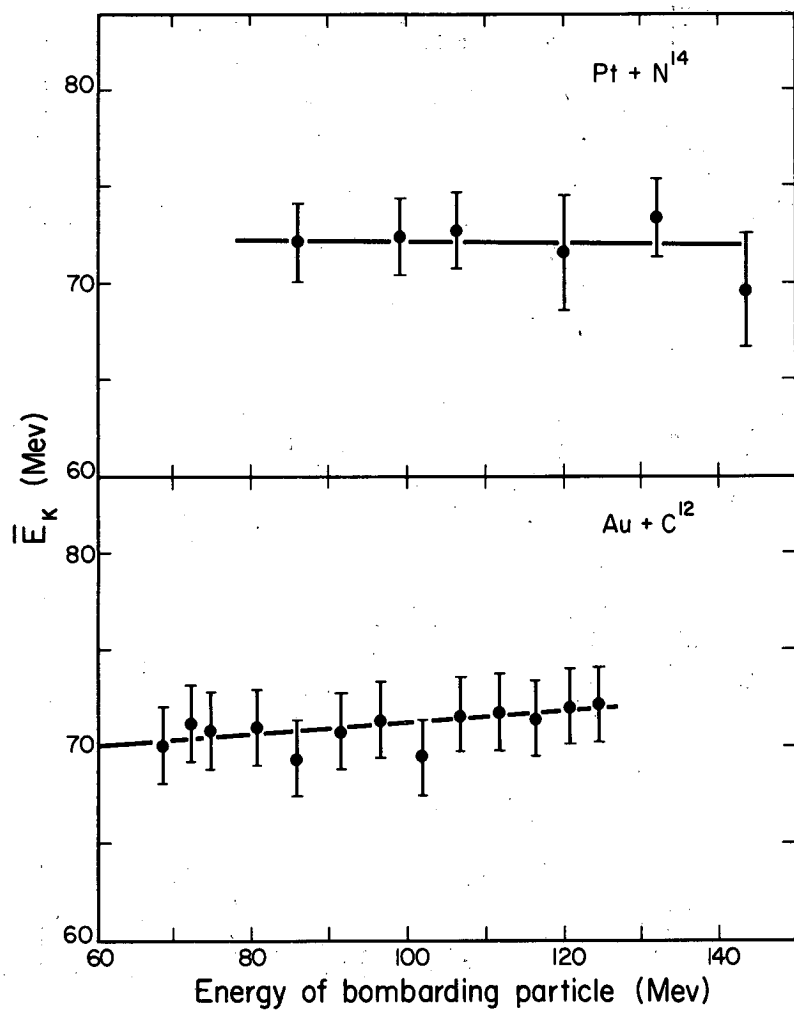
$$E_{\text{lab}}(90 \text{ deg}) = E_{\text{cm}}(1 - \eta^2), \quad (\text{II-13})$$

thus, the center-of-mass energy obtained from the laboratory energy is not very sensitive to the η value used. From the results of the data on kinetic energy vs laboratory angle, the η values corresponding to full momentum transfer have been used in this equation. The results of these experiments are shown in Fig. 23.

3. Angular Distributions

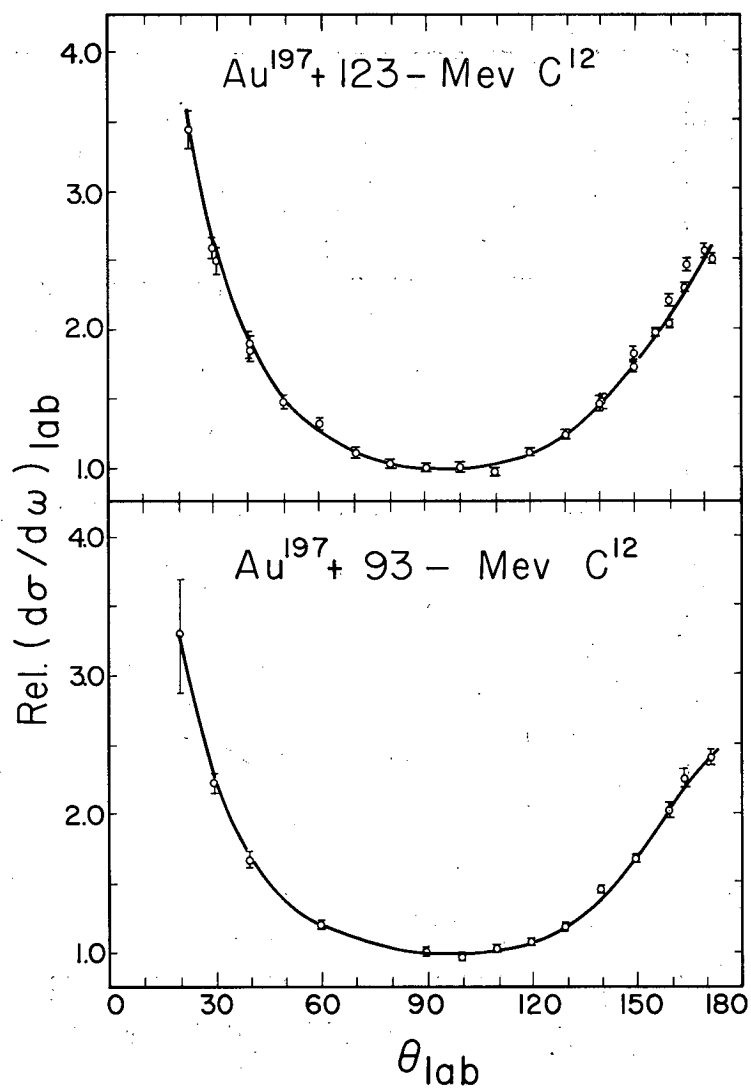
In order to obtain angular distributions of the fission fragments, the number of counts occurring in the fragment peaks of the kinetic-energy spectra were summed for each angle. At forward angles it was often necessary to perform a logarithmic subtraction of the background due to "pile-up" pulses in order to determine the shape of the low-energy tail of the fragment spectra. The total number of fragments was corrected for coincidence loss, and data at various angles were normalized to the same amount of beam. During each series of angular-distribution experiments, measurements were frequently taken at 90 deg to detect any changes in target thickness, absorber thickness, etc. The number of counts observed at each angle per unit beam was divided by the average number observed at 90 deg in order to normalize the angular distributions to unity at 90 deg. The resulting angular distributions in the laboratory system are shown in Figs. 24 and 25. The limits of error shown in these figures represent standard deviations in the number of counts plus estimated uncertainty in extrapolation of the low-energy tails of the fragment kinetic energy spectra. The limits of error on the points of the Au + 72.4-Mev C¹² distribution are rather large because the fission cross section is small at this energy; consequently, the number of fragments observed in a reasonable time of bombardment was much smaller than for the other systems.

As a first approximation, values of η^2 from the kinetic energy data were used to transform the laboratory system angular distributions into the center-of-mass system. (A summary of the η values used in the final transformations is given in Table II. The transformed angular distributions are shown in Figs. 32-36 below.)



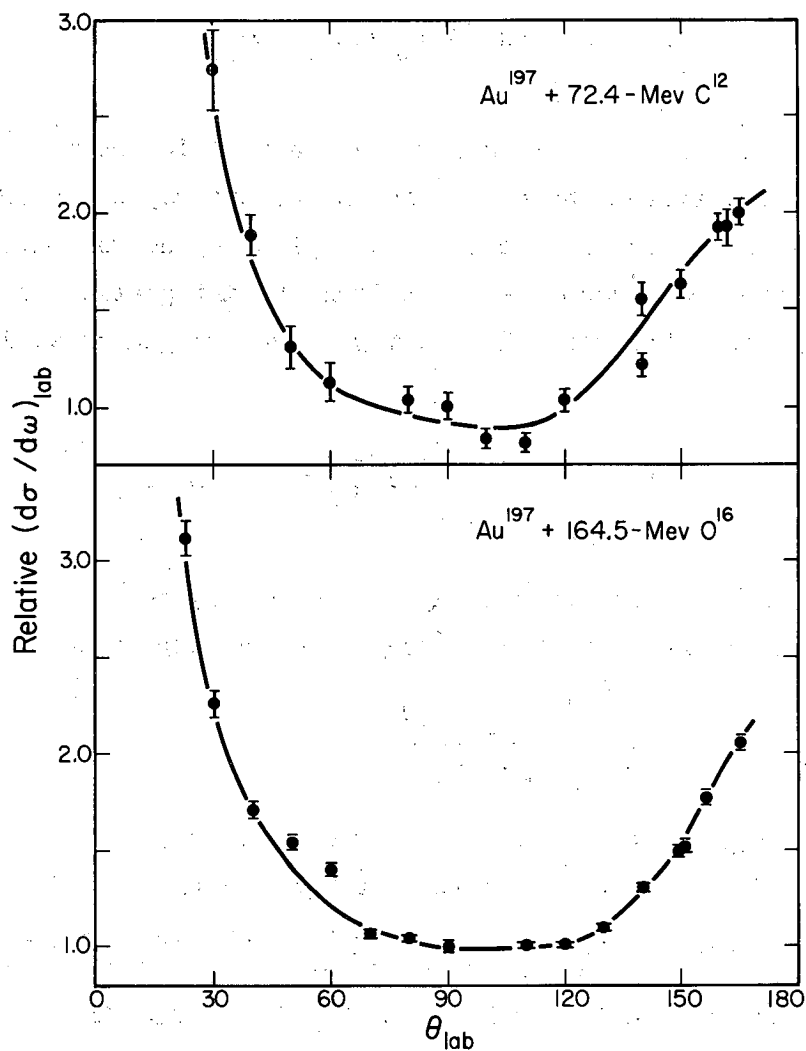
MU-19862

Fig. 23. Most probable kinetic energy per fragment in the center-of-mass system as a function of bombarding energy.



MU-18878

Fig. 24. Angular distribution of fission fragments in the laboratory system. Upper curve, $Au^{197} + 123-Mev C^{12}$; lower curve, $Au^{197} + 93-Mev C^{12}$.



MU-19864

Fig. 25. Fission fragment angular distributions in laboratory system. Upper curve: $\text{Au} + 72.4\text{-Mev C}^{12}$; lower curve: $\text{Au} + 164.5\text{ Mev O}^{16}$.

4. Total Fission Cross Sections

The total fission cross section is given by the expression

$$\sigma = 2\pi \int_0^{\pi} (d\sigma/d\omega)_{cm} \sin\theta_{cm} d\theta_{cm} \quad (II-14)$$

This expression is equally valid if all quantities on the right refer to the laboratory system; however, because of the rapid increase of the angular distributions at the forward angles in the laboratory system, it was considered more accurate to perform the integration in the center-of-mass system. Equation (II-14) may be rearranged to yield the expression

$$\sigma = 2\pi (d\sigma/d\omega)_{90^\circ} \int_0^{\pi} \frac{(d\sigma/d\omega)_{\theta}}{(d\sigma/d\omega)_{90^\circ}} \sin\theta d\theta, \quad (II-15)$$

where all angles refer to the center-of-mass system.

In operational terms, Eq. (II-15) may be expressed as

$$\sigma = \frac{2\pi (dN_{frag}/d\omega)_{90}}{2N_{tgt} N_{bp}} X, \quad (II-16)$$

where N_{tgt} = number of target atoms per cm^2 ,

N_{bp} = number of bombarding particles,

$(dN_{frag}/d\omega)_{90}$ = number of fragments per unit solid angle emitted at 90 deg in the center-of-mass system,

and X represents the integral of Eq. (II-15). The factor of 2 must be included in the denominator because two fragments are emitted per fission event.

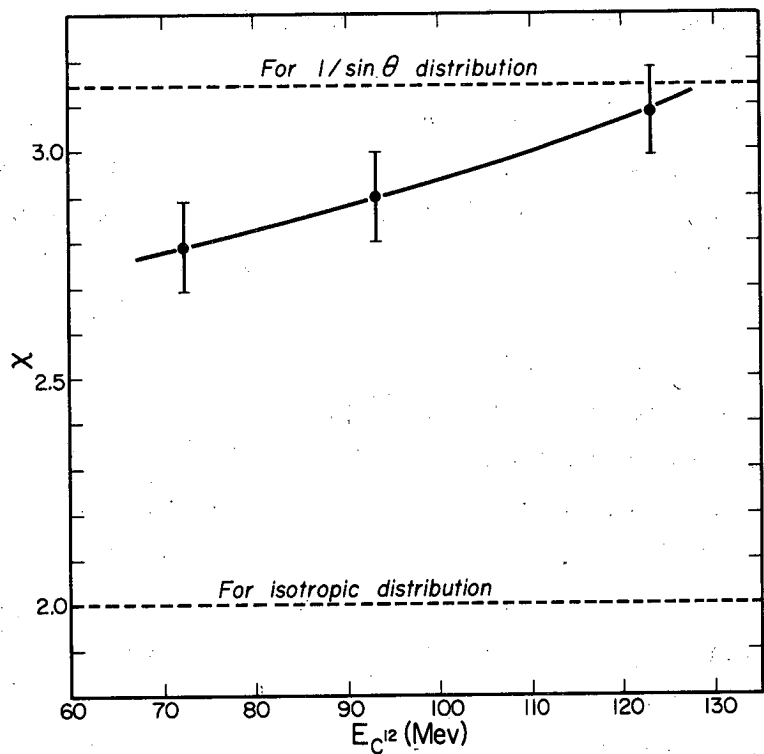
Values for $(dN_{frag}/d\omega)_{lab,90}$ as a function of bombarding-particle energy were obtained from experiments in the "T" assembly. In determining these values, it was necessary to calibrate the geometry of the gas scintillation chamber attached to the assembly. This was done by counting fission fragments from a Cf^{252} sample of known activity placed in the target position. A small correction was also necessary for the number of beam particles determined from the Faraday-cup readings. This

correction is discussed in Appendix A. Values for $(dN_{\text{frag}}/d\omega)_{\text{cm},90}$ were obtained from those for $(dN_{\text{frag}}/d\omega)_{\text{lab},90}$ by use of the angular distributions and the transformations.

For $\text{Au} + \text{C}^{12}$ fission, the integral X was evaluated at the three bombarding energies for which the angular distributions were obtained. A smooth curve was drawn through the points of the center-of-mass angular distributions. At each 5-deg interval the value of this curve was multiplied by $\sin \theta_{\text{cm}}$ and the resulting curve was graphically integrated. The three values of X thus obtained are plotted as a function of carbon particle energy in Fig. 26. A smooth variation of X between the measured points is assumed. It should be noted that the value of X is rather insensitive to small errors in the angular distributions, as it varies only between a value of 2 for isotropic fission and a value of π for a $1/\sin \theta$ angular distribution.

No angular distributions were obtained in the $\text{Pt} + \text{N}^{14}$ system. It was felt that the added complication of the many isotopes of platinum would make the interpretation of such angular distributions less clear-cut. The values of X for this system have been estimated by assuming that they are equal to the values obtained in the $\text{Au} + \text{C}^{12}$ system when the compound nuclei in the two systems have the same average angular momenta.

The fission excitation functions resulting from this analysis of the data are shown in Fig. 27. For the $\text{Au} + \text{C}^{12}$ cross sections it should be noted that although Faraday-cup, geometry, and target-thickness measurements were made, any errors in these determinations cancel out because all these data are related to the elastic-scattering cross sections of $\text{Au} + 72.4\text{-Mev } \text{C}^{12}$ which were used for calibration of the Faraday cup (cf. Appendix A). Thus the magnitudes of the $\text{Au} + \text{C}^{12}$ fission cross sections are based on the assumption that at small angles the elastic-scattering cross sections of $\text{Au} + 72.4\text{-Mev } \text{C}^{12}$ are equal to the values calculated from the Rutherford equation. The limits of error on these points thus represent standard deviations of the number of fragments observed at 90 deg, and the number of elastically scattered particles



MU-20132

Fig. 26. Value of $\chi = \left(\int_0^\pi (d\sigma/d\omega)_\theta / ((d\sigma/d\omega)_{90^\circ} \sin\theta d\theta)_{cm} \right)$ as a function of bombarding energy for fission of $Au + C^{12}$.

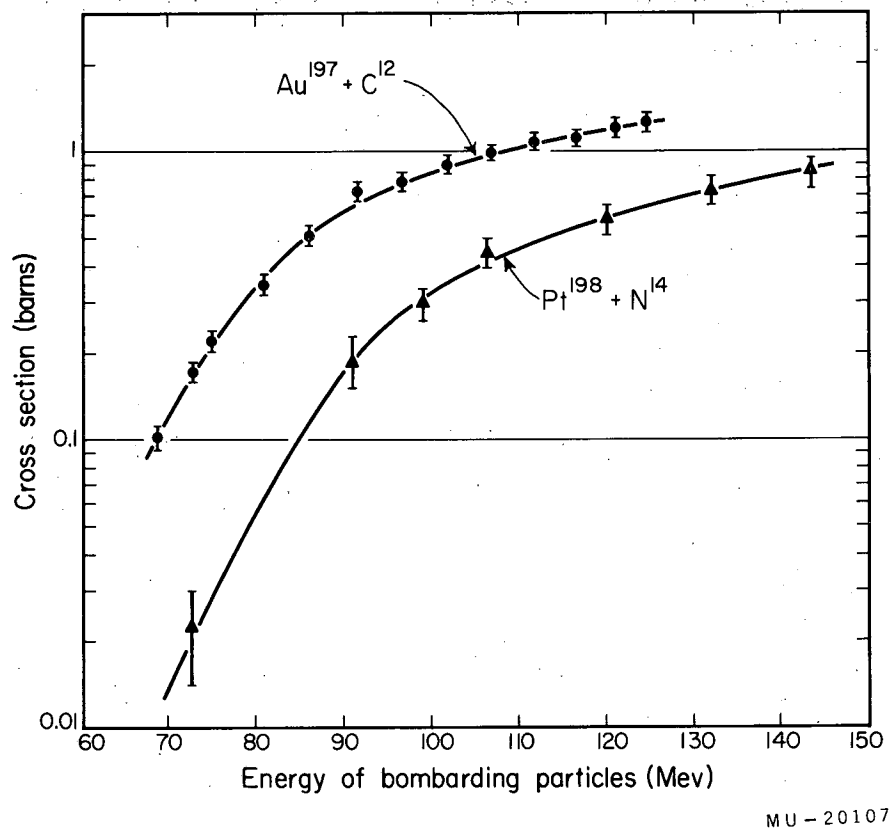


Fig. 27. Fission-excitation functions for Au + C¹² and Pt + N¹⁴.

observed at small angles plus estimated uncertainty in the value of χ . For Pt + N¹⁴, it was not possible to relate the fission cross sections to elastic-scattering cross sections because all counting was done at 90 deg. Thus it is necessary to include uncertainty in the target thickness. This may be rather large, as an electroplated target was used in these experiments.

III. SPALLATION EXPERIMENTS

A. Introduction

In this section, experiments are described in which the cross sections for various $\text{Pt}(\text{N}^{14}, \text{xn})\text{At}$ reactions have been determined. Natural platinum consists of 7.2% Pt^{198} , 25.4% Pt^{196} , 33.7% Pt^{195} , 32.8% Pt^{194} , 0.78% Pt^{192} , and 0.012% Pt^{190} . In order to obtain individual cross sections for the reactions involving the various platinum isotopes, two types of experiments have been performed.

In one series of experiments, a thin, natural-platinum target on 0.25-mil Al backing foil was bombarded with N^{14} at various energies. The target was mounted with the platinum on the backside of the aluminum foil. By means of the recoil technique previously described by several authors,⁷³⁻⁷⁵ the astatine product nuclei were caught in another 0.25-mil Al foil behind the target. Owing to conservation of the linear momentum of the incoming beam particles, the recoiling nuclei have rather large energies (~4 to 9 Mev) and ranges in platinum.^{76,77} In preliminary experiments it was found that with targets up to about $100 \mu\text{g}/\text{cm}^2$ thick, approx 99% of the alpha activity produced was caught in the recoil catcher. By means of the tracer method described below, the amount of one or two of the predominant activities produced was determined as a function of bombarding energy.

In the other type of experiments, ratios of various activities produced were determined as functions of bombarding energy for the natural isotopic mixture and platinum enriched in Pt^{198} (60.95% Pt^{198} , 26.47% Pt^{196} , 8.97% Pt^{195} , 3.57% Pt^{194} , 0.042% Pt^{192} , and 0.012% Pt^{190}). Combining these pieces of information with the isotopic compositions of the targets, it was possible to determine some of the $\text{Pt}(\text{N}^{14}, \text{xn})\text{At}$ cross sections.

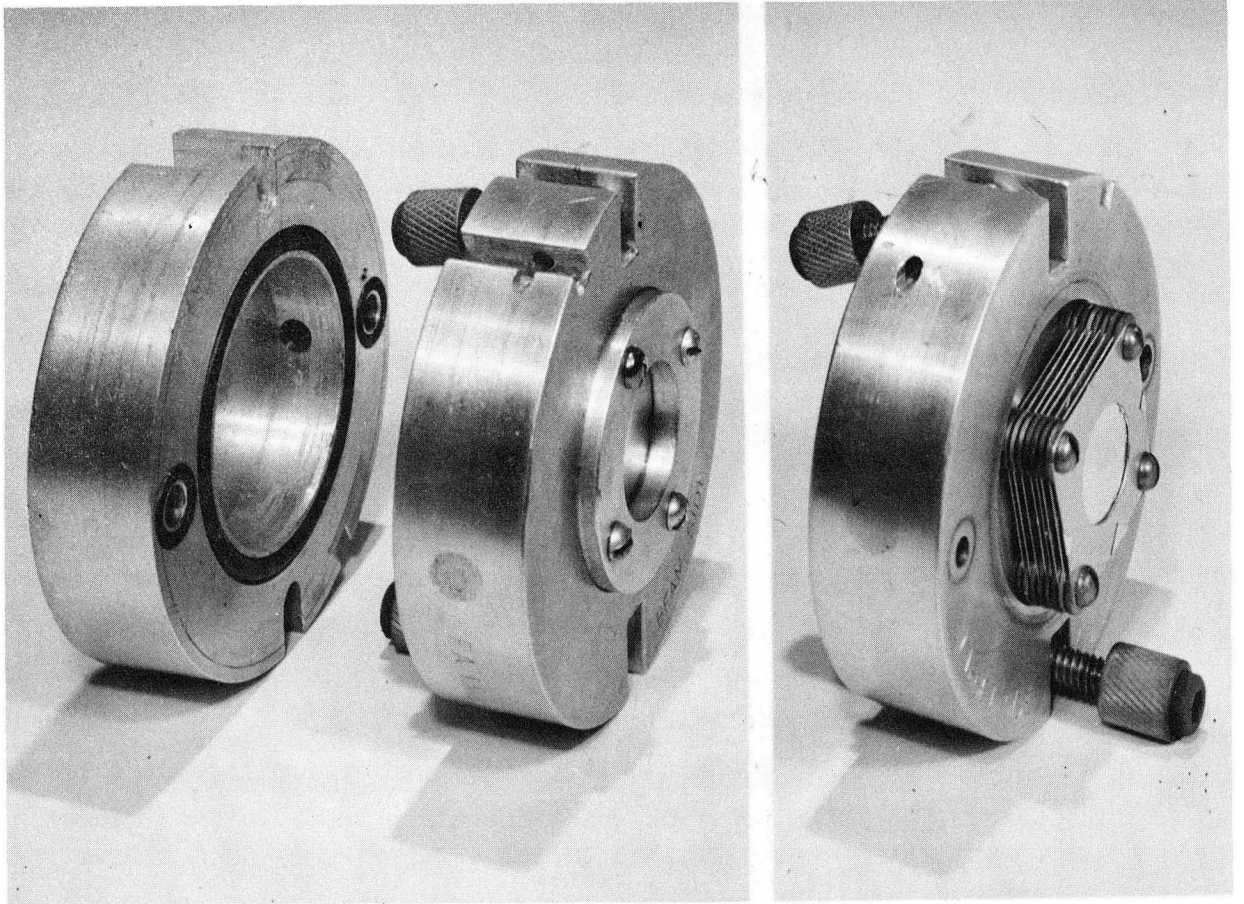
B. Experimental Procedures

1. Target Preparation and Assemblies

The target used in the quantitative experiments was prepared by vaporizing natural platinum from a hot tungsten filament. During the vaporization, the 0.25-mil Al backing foil was masked so that the coated area was well defined. The thickness of the deposit ($60 \mu\text{g}/\text{cm}^2$) was determined by weighing the backing foil before and after vaporization. The target was mounted on a probe and placed in a "T" assembly similar to that shown in Fig. 43 of Appendix A. The recoil catchers were also mounted on probes and positioned behind the target. The energy of the bombarding particle was varied by placing aluminum degrading foils in the beam path ahead of the "T" assembly. Energies were determined from the range-energy data of Sikkeland⁷⁸ and Roll and Steigert.⁷⁹

Thicker targets (about 200 to $300 \mu\text{g}/\text{cm}^2$) were used for the ratio experiments in order to produce more activity. The targets were made by the standard electroplating procedure for platinum.⁸⁰ Metallic platinum was dissolved in aqua regia and evaporated nearly to dryness several times in the presence of excess HCl. This destroyed most of the nitrate and yielded H_2PtCl_6 . The chloroplatinic acid was added to an aqueous solution of diammonium and disodium phosphates. The resulting plating solution was boiled for several hours before being added to the electroplating cell. Metallic platinum was plated onto a 0.1-mil Ag foil which formed the base of the plating cell. During the plating, which normally lasted about 20 min, the voltage was kept at approx 2 v, with the current at approx $0.001 \text{ amp}/\text{cm}^2$. The plating solution was stirred by bubbling air, and kept near the boiling point by a heating coil around the cell. Thicknesses were determined by weighing the foil before and after plating. The sample of metallic platinum enriched in Pt^{198} was obtained from Oak Ridge National Laboratory.

The targets were attached to stainless steel holders which could be mounted in the target portion of the TAG assembly (Target assembly, general) shown in Fig. 28. In order to preserve the targets, the recoil method was also used in these experiments. The 0.25-mil Al catcher foils



ZN-2456

Fig. 28. Hilac TAG (Target assembly, general) target holder.

were mounted on holders and placed between the targets. Spacers were placed between all the foils to allow helium gas to circulate between them and minimize heating. The entire target assembly, including the absorbers, served as the Faraday cup. No absolute cross sections were obtained with this arrangement, as the absolute beam-current measurements were not considered reliable.

2. Chemical Separations

Preliminary experiments showed that no At^{211} was produced in the $\text{Pt} + \text{N}^{14}$ bombardments, thus making it possible to use At^{211} as a tracer when quantitative yields were desired. The At^{211} was produced free of other detectable alpha activity by bombarding Bi^{209} with 28-Mev He^4 ions at the Crocker 60 inch cyclotron.³⁶ In order to have the astatine tracer in the same chemical environment as the Hilac-produced activity, the At^{211} atoms were also caught in an aluminum recoil-catching foil. Prior to the heavy-ion bombardments, the recoil catcher was cut up and the alpha particles being emitted from each sample counted to obtain the desired amounts of tracer activity. Following the heavy-ion bombardment, the recoil catcher containing the $\text{Pt} + \text{N}^{14}$ products and a plate containing a known amount of tracer activity were simultaneously dissolved in 8 M HCl. The astatine was separated from other activities by extraction into DIPE (di-isopropyl ether). The DIPE fraction was transferred to a platinum counting disk and evaporated to dryness under a heat lamp. The resulting sample was placed in an alpha grid chamber and pulse analyzed to determine the amounts of principal alpha activities relative to that of the At^{211} tracer.

It is felt that the above procedure is a reliable method for determining the chemical yield of the astatine. As nearly as possible, the tracer and heavy-ion-produced activities have received the same treatment throughout the experiments. The only obvious difference in the environments of the two astatine fractions is the depth of the deposits in the aluminum foils. The recoils produced by 28-Mev He^4 bombardment receive less momentum, and thus are deposited closer to the surface than the recoils from $\text{Pt} + (\geq 60\text{-Mev}) \text{N}^{14}$ reactions.

In experiments in which only the ratios of activities were determined, the astatine fraction was separated by a double volatilization method.⁸¹ The aluminum recoil catcher was cut from its holder, crumpled up, and dropped into a quartz cup similar to that described by Hollander.⁸² The cup was heated with a methane-oxygen blowtorch, melting the aluminum and volatilizing the astatine. The astatine was condensed on an aluminum disk attached to a water-cooled "cold finger." The aluminum disk was then detached from the cold finger, placed in another quartz cup, and gently warmed with a microburner. In this step, the astatine was condensed on a platinum counting plate attached to the cold finger. This double volatilization yielded astatine free of other alpha-emitting products. The most serious contamination could come from the large amounts of polonium present. However, tracer experiments showed that about 50% of the polonium comes over in the first step, but less than 0.02% in the second. The yield of astatine from the two steps was about 50 to 75%. There was no evidence for loss of astatine from the platinum plate during counting, but the longest-lived isotope studied, aside from the tracer, was At²⁰⁷, which has a 1.8-hr half life. Samples produced by this method were essentially mass-free and quite good for alpha pulse analysis.

Since the samples produced by the use of the DIPE were rather thick and gave poorly resolved peaks upon alpha pulse analysis, an attempt was made to adapt the double-volatilization method for quantitative yield determinations. The astatine from the recoil catcher and plate containing the tracer was vaporized simultaneously and the results were compared with those obtained from the DIPE chemistry. The results of the two methods generally agreed within 15%, but the ratio of tracer activity to activity in the recoil catcher was uniformly higher when the vaporization method was used. This may have been due to the greater depth of deposition of the heavy-ion-produced recoils. In any case, the chemical method was considered more reliable.

3. Counting Procedures

The only absolute counting was that of the plates containing At^{211} tracer. This was done in an ionization chamber of 50% geometry.

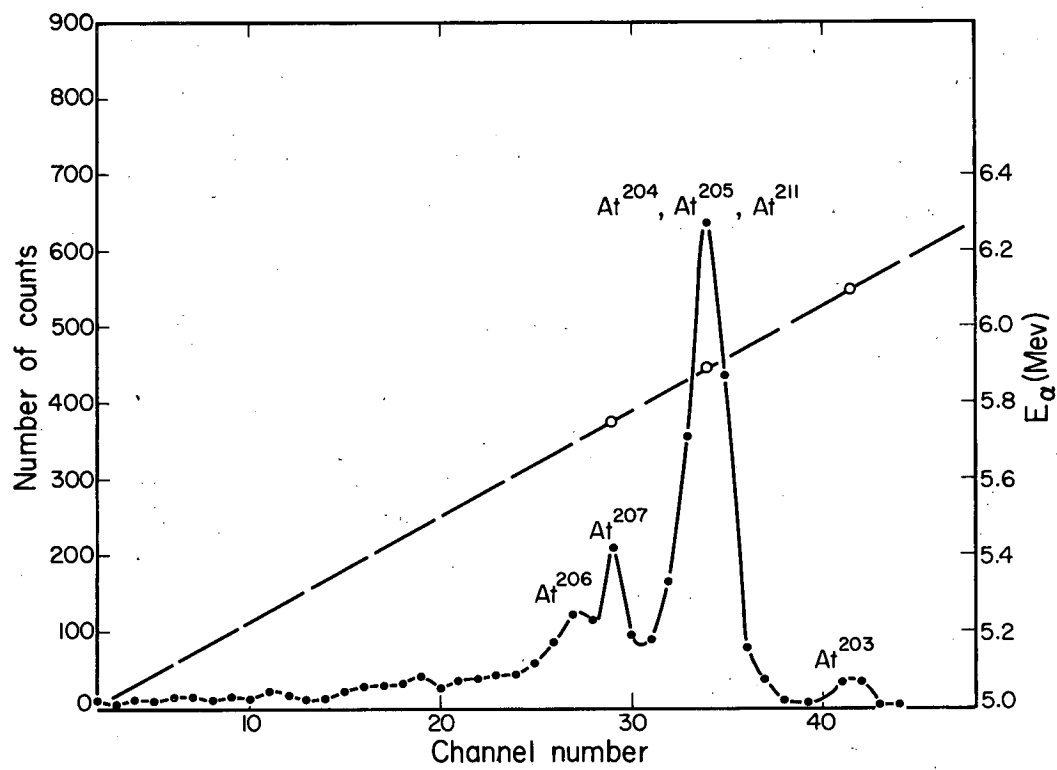
For determining the ratio of various activities to At^{211} tracer, the samples produced by the DIPE chemistry method were pulse analyzed in an alpha grid chamber. An example of the results of pulse analysis is shown in Fig. 29. Note that the 5.89-Mev alpha group of At^{211} is obscured by pulses from At^{205} and At^{204} alpha particles. For this reason, the number of 7.3-Mev alpha particles from Po^{211} (the electron-capture daughter of At^{211}) was used for determination of the tracer activity. The intensity of the At^{205} and At^{204} alpha groups was determined by subtracting the number of alpha particles due to At^{211} from the peak at approx 5.9 Mev by use of the α/EC branching ratio of At^{211} .

In the ratio experiments, pulse analysis was used to determine the relative amounts of various activities produced. Examples of such pulse analyses are shown in Figs. 30 and 31. Note that the resolution of the various peaks is considerably better with the thin samples produced by means of the double vaporization method than with those resulting from the extraction chemistry.

By pulse analysis alone, it was not possible to separate the 5.69-Mev alpha particles of At^{206} from the 5.75-Mev group of At^{207} , nor those of 5.95-Mev energy due to At^{204} from 5.89-Mev particles of At^{205} . Fortunately the half lives are sufficiently different that those activities could be separated by following the peaks as a function of time and resolving the decay curves. The starting times of the counts were accurately determined from a stopwatch started at the end of bombardment. In most cases, the initial count was started at 6 to 7 min after the end of bombardment.

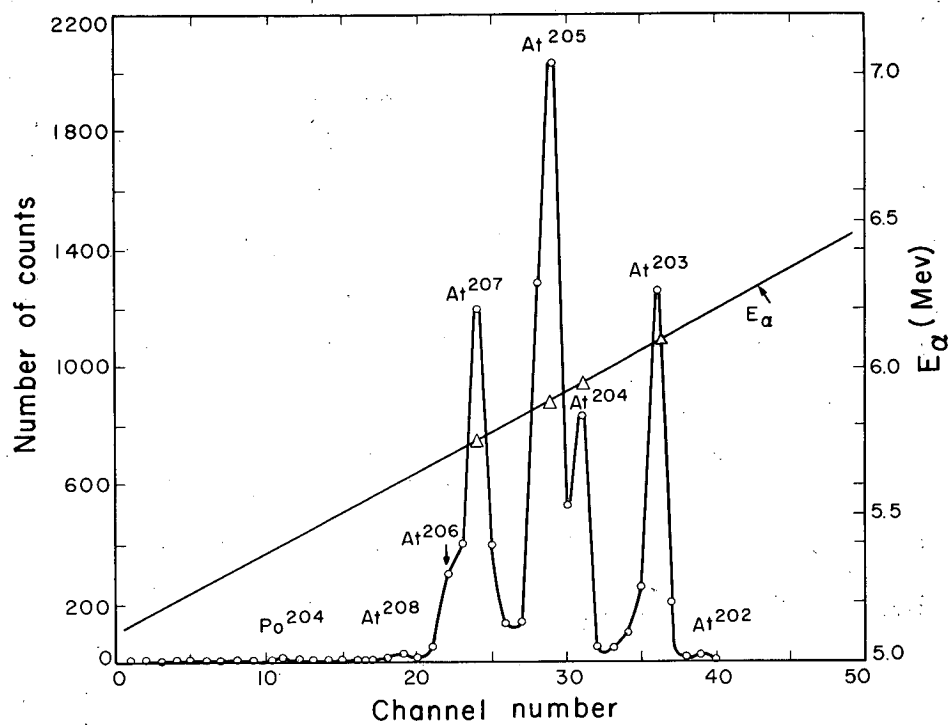
4. Treatment of the Data

The amounts of each activity observed were plotted as a function of time and extrapolated back to the end of bombardment. The half lives observed were, within experimental error, usually those obtained in previous work or those determined in this study.



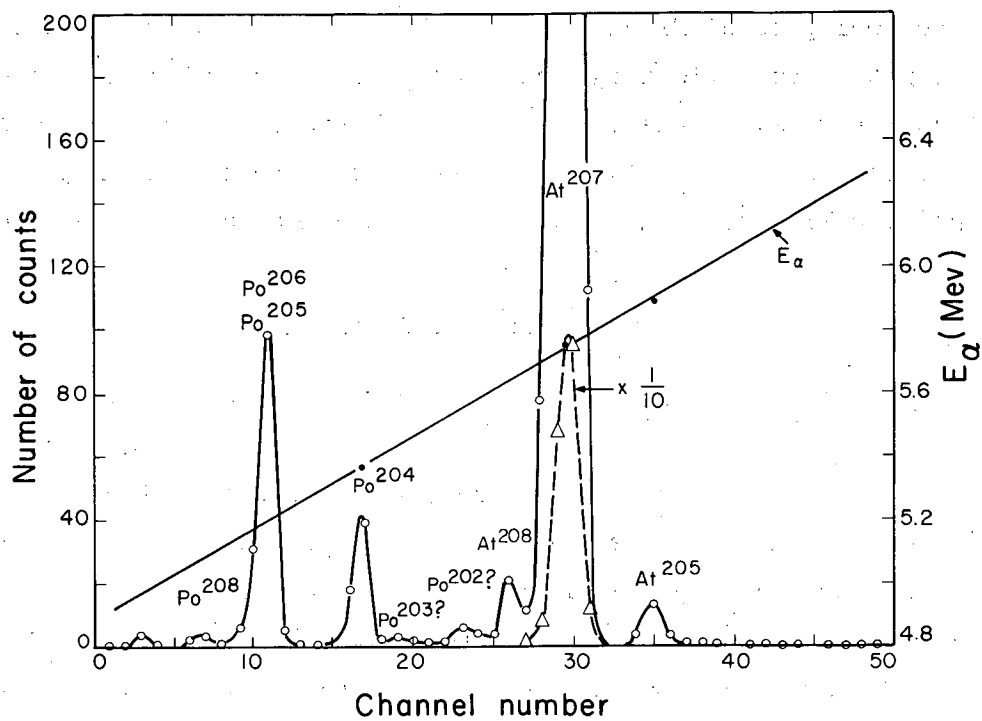
MU-20115

Fig. 29. Alpha pulse analysis of a sample produced by DIPE method.



MU-19025

Fig. 30. Alpha-particle pulse analysis of a sample produced by double-volatilization method (3 min after end of bombardment).



MU-19027

Fig. 31. Alpha-particle pulse analysis of sample used in Fig. 30 (320 min after end of bombardment).

Corrections for decay during bombardment and counting were made by using the equations for growth and decay of activity.⁸³ A chart of beam current as a function of time was used for making the decay-during-bombardment corrections. A constant was generally sought, but often not obtained owing to difficulties with the accelerator. In such cases, the beam charts were divided into portions, in which the beam was relatively constant, for making the corrections. Corrections were also applied for the electron-capture decay of the astatine isotopes. Half lives and α /EC branching ratios used in the calculations are discussed in Appendix C.

C. Experimental Results

The absolute yields of one or two of the principal activities (At^{203} , At^{205} , or At^{207}) produced by nitrogen bombardments of the vaporized, natural platinum target were obtained at various energies by means of the tracer experiments. These data, combined with the ratios of various activities produced by bombardments of the enriched- Pt^{198} and natural-platinum targets, were used to obtain cross sections of the various reactions. In several cases, where the same product resulted from reactions involving more than one platinum isotope, it was necessary to use a set of linear equations to obtain the individual cross sections.

One important assumption should be noted. Since targets of only two different isotopic compositions were used, it was necessary to have at each energy one product that is produced by only one reaction, in order to obtain the individual cross sections. At the lower energies, it has been assumed that At^{207} is produced only by the $(\text{N}^{14}, 5n)$ reaction on Pt^{198} . This implies that the $\text{Pt}^{196}(\text{N}^{14}, 3n)\text{At}^{207}$ reaction cross section is negligible. It was not possible to test this assumption directly. However, very careful observations of the alpha-particle spectra from samples produced in low-energy bombardments gave no evidence for production of At^{209} , the product of the $\text{Pt}^{198}(\text{N}^{14}, 3n)$ reaction. This result, plus the observation of very small $(\text{N}^{14}, 4n)$ reaction cross sections, is taken as evidence for validity of the assumption. At the highest energies (about 100 Mev), it was assumed that most of the At^{205} resulted from the

$\text{Pt}^{198}(\text{N}^{14}, 7\text{n})$ reaction. A small correction was made for the amount from the $\text{Pt}^{196}(\text{N}^{14}, 5\text{n})$ reaction by extrapolating the high-energy tail of the excitation function.

The excitation functions obtained by these procedures are shown in Figs. 38 and 39 of Section IV. Since the data from the three types of experiments were not usually obtained at exactly the same bombarding energies, smooth curves were drawn through the experimental points corresponding to the absolute yields and ratios of isotopes produced as functions of energy. Thus, for reactions whose cross sections are given at regularly spaced energy intervals, this is not meant to imply that cross sections were determined at exactly those energies, but that the simultaneous equations were solved there. Also, in these cases, scatter in the experimental points was necessarily removed by the smoothing process. The limits of error given for these cross sections represent estimated uncertainties in the positions of the smoothed curves. These estimates, as well as the limits of error given for the other cross-section points, include uncertainties in the counting rates of the radiations from the product nuclei, target thickness, and decay during bombardment corrections. Uncertainty in the values used for the α/EC branching ratios have not been included.

In addition to the experimental results given, data were also obtained on the production of At^{203} and At^{202} . However, these products result from so many different reactions that occur with large probability that it was not possible to determine the individual cross sections. The data for the $\text{Pt}^{198}(\text{N}^{14}, 8\text{n})\text{At}^{204}$ reaction are also somewhat poor owing to this effect, and possibly the limits of error should be greater than indicated.

IV. INTERPRETATION OF RESULTS

A. Angular Distributions

The general approach to the interpretation of angular distributions of fission fragments was first proposed by Bohr.²³ According to his model, for excitation energies only slightly above the fission barrier, the nucleus goes over the saddle-pass "cold"; that is, most of the excitation energy is expended in potential energy of deformation towards fission. Thus, the spectrum of energy levels of the highly deformed nuclei at the saddle pass should be similar to those of stably deformed nuclei at energies near their ground states. Bohr further assumes that the nuclei retain axial symmetry throughout the deformation, and that the fragments are emitted in the direction of the symmetry axis. The fragment angular distributions are therefore given by the distributions of the orientations of the symmetry axes with respect to the beam.

Angular distributions based on the Bohr theory have been worked out quantitatively and extended to higher energies by a number of authors.^{24,25,84-86} The treatments by Griffin²⁵ and Halpern and Strutinski²⁴ are most applicable to the cases studied in this work. Since these treatments are similar in most details, the Halpern and Strutinski method will be described in detail, with the differences introduced by Griffin's calculations pointed out below.

According to the Halpern and Strutinski treatment, the fissioning nucleus is characterized by three quantum numbers: I , the total angular momentum; K , the projection of I on the nuclear symmetry axis; and M , the component of I in the beam direction. For fission induced by high-energy heavy ions, I is approximately equal to the orbital angular momentum of the bombarding particle, ℓ , which is perpendicular to the beam direction. Since any target and bombarding-particle spins are small compared with ℓ , it is quite reasonable to assume $M = 0$; i.e., $\vec{I} = \vec{\ell}$, $\vec{I}$ is perpendicular to the beam direction. With these assumptions, the angular distribution for a given I and K is

$$W_{I,K} = \frac{2I}{4\pi} (I^2 \sin^2 \theta - K^2)^{-1/2}, \quad (\text{IV-1})$$

where θ is the center-of-mass angle between the fragment direction and the beam.

Based on statistical arguments, the probability distribution of K is assumed to be

$$F(K) \propto \exp(-K^2/2K_0^2), \quad (\text{IV-2})$$

where K_0^2 is the mean value of K^2 . The angular distribution of Eq. (IV-1) is then multiplied by the weighting function (Eq. (IV-2)) and integrated over K to obtain the angular distribution for given values of K_0^2 and I :

$$W_{I,K_0} = \frac{2I}{4\pi^2} \frac{\int_0^{I \sin \theta} dK \exp(-K^2/2K_0^2) (I^2 \sin^2 \theta - K^2)^{-1/2}}{\int_0^I dK \exp(-K^2/2K_0^2)}, \quad (\text{IV-3})$$

$$= \frac{2}{\pi} \frac{N}{2\pi} \frac{I}{2K_0} \exp\left(\frac{-I^2 \sin^2 \theta}{4K_0^2}\right) J_0\left(\frac{iI^2 \sin^2 \theta}{4K_0^2}\right), \quad (\text{IV-4})$$

where J_0 is the zeroth-order Bessel function, and N is a normalizing factor. This distribution is then integrated over I from zero up to the maximum value I_m , assuming the probability of I^2 is constant. The resulting angular distributions, characterized by the parameter p ($= I_m^2/4K_0^2$) behave as $1/\sin\theta$ in the region near 90 deg, falling below it as 0 and 180 deg are approached. The higher the value of p is, the longer the angular distribution follows $1/\sin\theta$.

Probably the least certain part of the treatment involves the choice of K_0^2 as a function of excitation energy, E_{ex} , and the height of the fission barrier, E_f . At high excitation energies in excess of the fission barrier, statistical theory predicts

$$K_0^2 \propto (E_{ex} - E_f)^{1/2}. \quad (\text{IV-5})$$

The relationship at low energies has not been clearly established. Halpern and Strutinski have obtained the function empirically from the experimental anisotropies of neutron fission of Th^{232} at various energies.⁸⁷ In this way they find K_0^2 to be approximately proportional

to $(E_{\text{ex}} - E_f)$ at low energies. At high excitation energies, the proportionality constant of Eq. (IV-5) was determined from the angular distribution of fragments from fission of Np^{237} with 43-Mev alpha particles.⁸⁸

Griffin's treatment of angular distributions is very similar to that by Halpern and Strutinski. However, he assumes

$$F(K) \propto K_{\text{max}} - K. \quad (\text{IV-6})$$

The angular distributions, characterized by $r(=\bar{I}/\bar{K})$ approximately follow $1/\sin\theta$ near 90 deg, but approach 0 and 180 deg linearly (illustrated below). At low energies, Griffin obtained $\bar{K}$ as a function of $(E_{\text{ex}} - E_f)$, from the experimental anisotropy of neutron fission of Pu^{239} at various energies.⁸⁹ This results in $\bar{K}$ approximately proportional to $(E_{\text{ex}} - E_f)$ rather than to $(E_{\text{ex}} - E_f)^{1/2}$ as noted above. At higher energies he also predicts the $1/4$ -power dependence, but does not indicate the energy at which the change in functions should occur.

The laboratory-system angular distributions obtained in this work were transformed to the center-of-mass system as discussed above, and are shown in Figs. 32-36. The most sensitive check on the ability of the theoretical angular distributions to fit the experimental points is provided by the data from 93-Mev C^{12} fission of gold. Figures 32 and 33 show the experimental angular distributions along with curves predicted by Halpern and Strutinski and by Griffin, respectively, using the indicated parameters. Although, in agreement with Halpern and Strutinski, the experimental points indicate some curvature near 0 and 180 deg, the statistics in none of the angular distributions are good enough to rule out the linear shape predicted by Griffin. Also, in every case, there are indications that the experimental points lie above the $1/\sin\theta$ curve in the regions approx 20 to 80 deg and approx 100 to 160 deg. This is in agreement with Griffin's predicted curves, which rise above the $1/\sin\theta$ function in these regions, for high values of r . Experimentally, this result is in very good agreement with the data of Viola,⁹⁰ who has determined angular distributions by a gross beta-counting method similar to

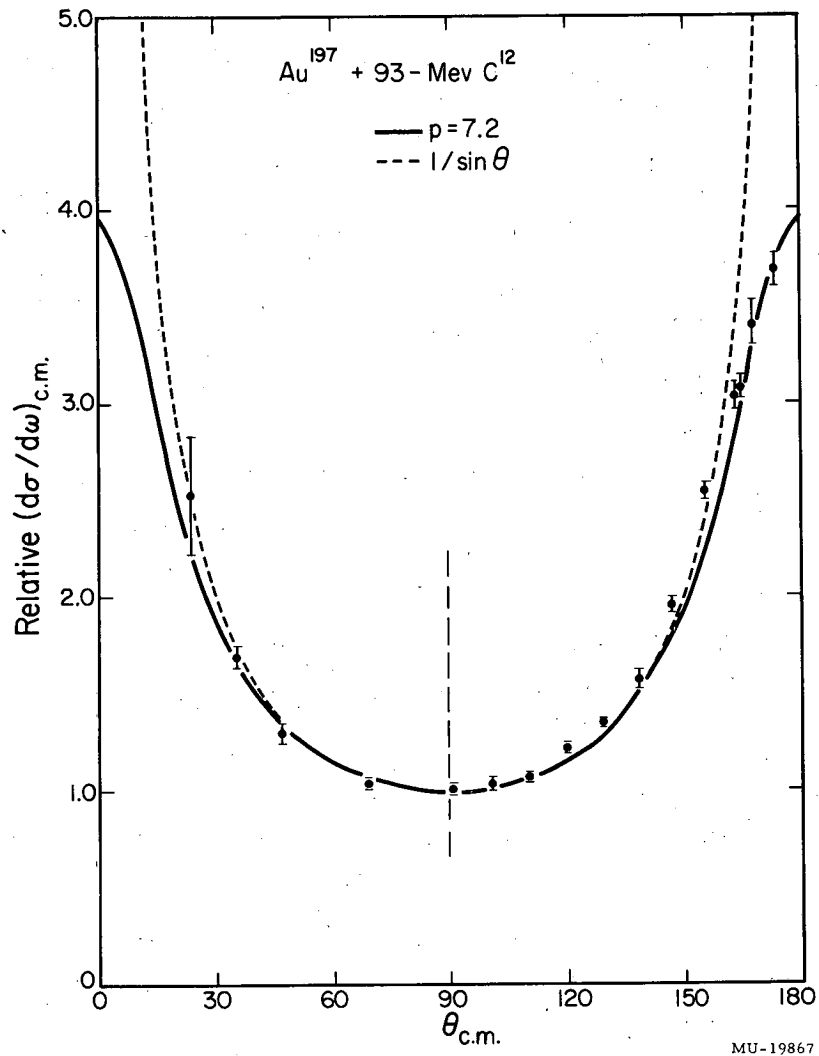


Fig. 32. Angular distribution (c.m. system) of fragments from fission of Au with 93.3-Mev C^{12} . Heavy curve: Halpern and Strutinski theoretical curve with $p = 7.2$.

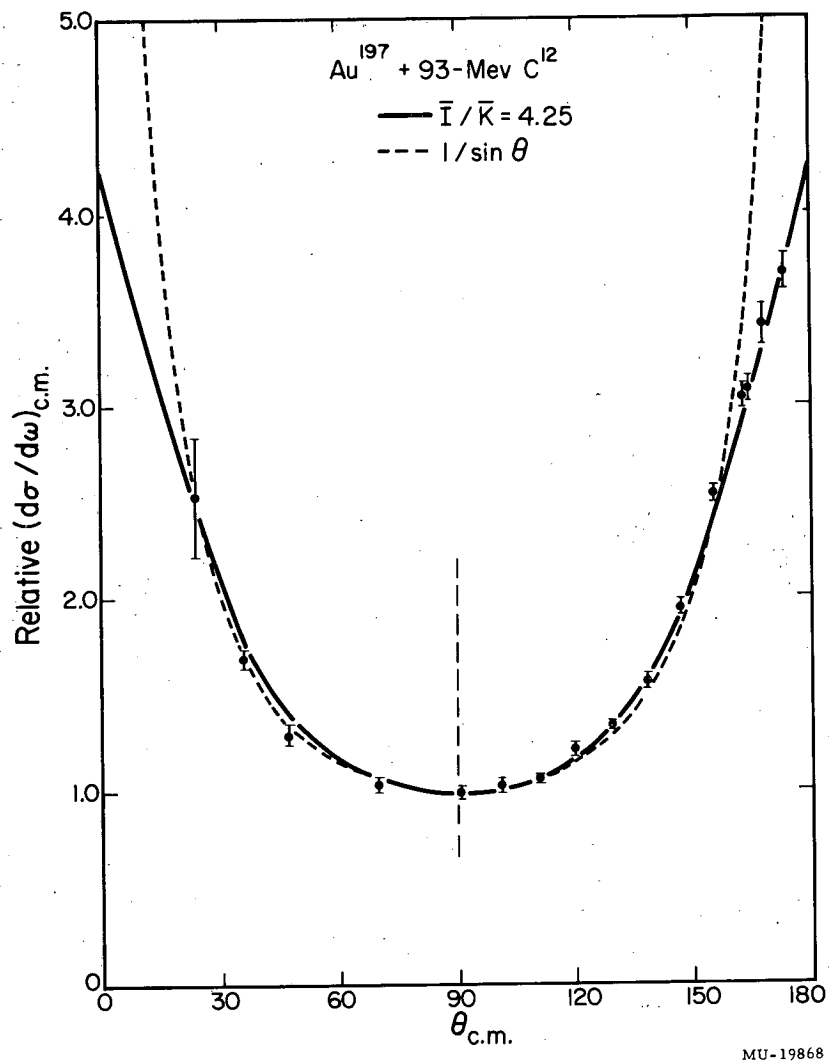


Fig. 33. Angular distribution (c.m. system) of fragments from fission of Au with 93.3-Mev C^{12} . Heavy curve: Griffin theoretical curve with $\bar{I}/\bar{K} = 4.25$.

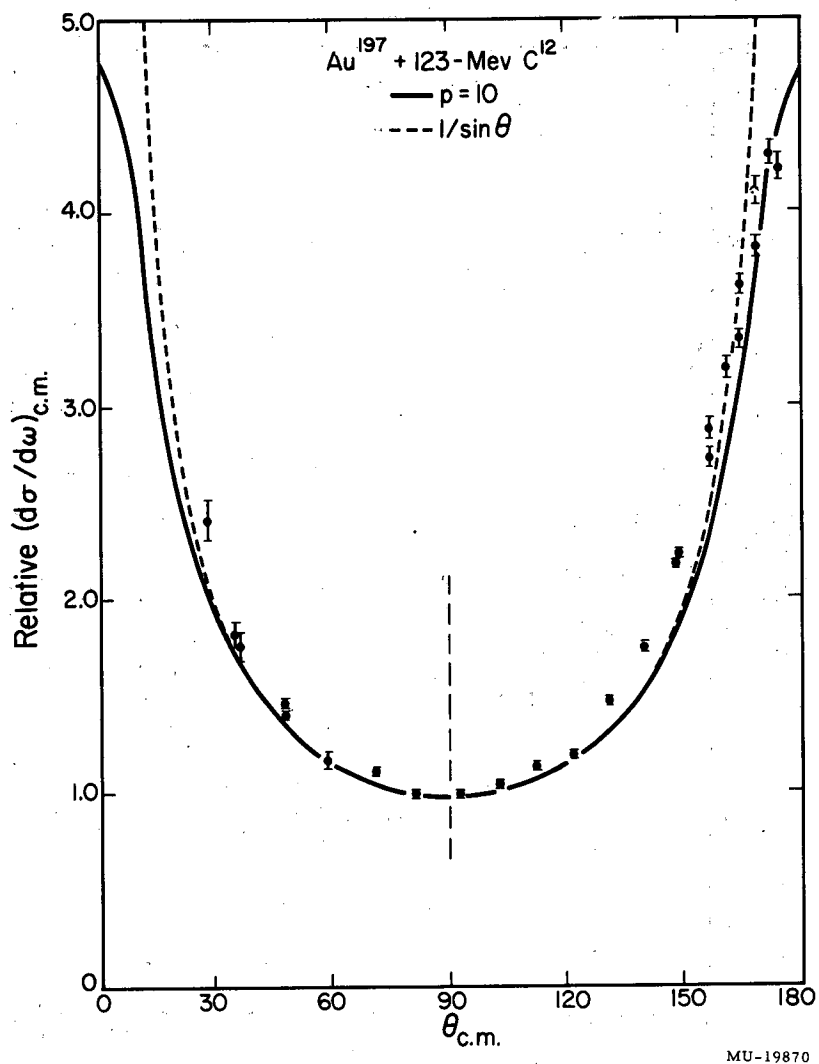


Fig. 34. Angular distribution (c.m. system) of fragments from fission of Au with 123.3-MeV C^{12} . Heavy curve: Halpern and Strutinski theoretical curve with $p = 10$.

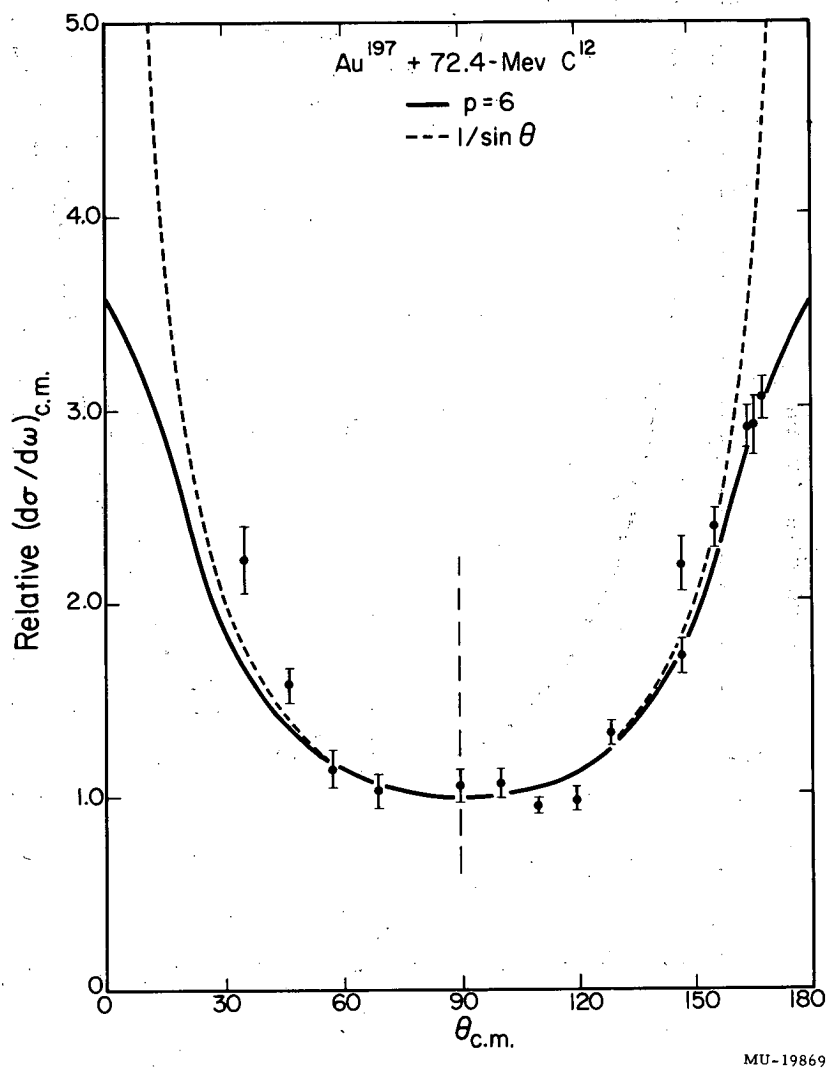


Fig. 35. Angular distribution (c.m. system) of fragments from fission of Au with 72.4-Mev C^{12} . Heavy curve: Halpern and Strutinski theoretical curve with $p = 6$.

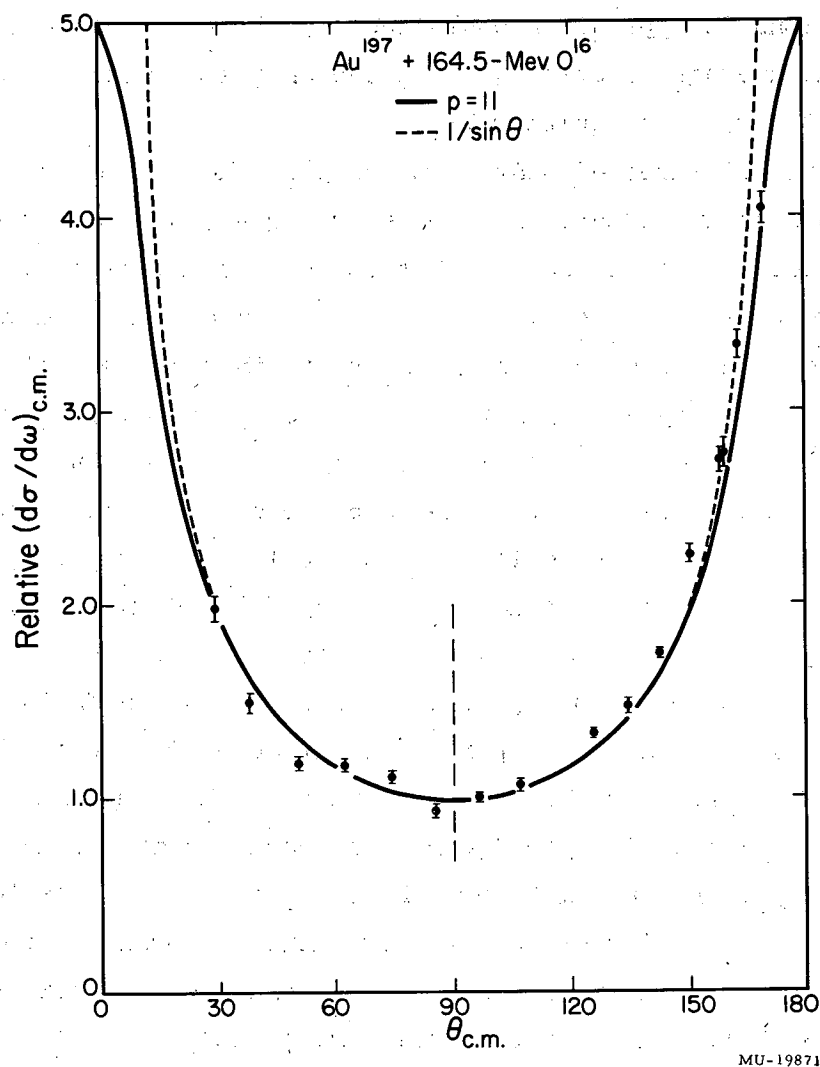


Fig. 36. Angular distribution (c.m.system) of fragments from fission of Au with 164.5-Mev O^{16} . Heavy curve: Halpern and Strutinski theoretical curve with $p = 11$.

that used by Coffin and Halpern.⁸⁸ It was first suspected that such an effect might arise in transformation of the laboratory-system angular distributions into the center-of-mass system by using a single value of η , whereas there is actually a distribution of η values. However, the calculations in Appendix B would indicate that such a transformation, using the average value of η^2 , reproduces the assumed center-of-mass angular distribution quite well.

It is not expected that scattering of the fragments could have produced such an effect. At the laboratory-system angles in the backward direction, corresponding to the center-of-mass-system angles at which the effect is observed, the cross section varies slowly with angle. The targets in all experiments were quite thin ($\leq 200 \mu\text{g}/\text{cm}^2 \text{ Au}$). Thus, small-angle scattering is not expected to be serious. If the large-angle scattering mentioned by Coffin and Halpern occurred, the scattered fragments would have had low energies and would either have been stopped in the detector window or registered in the low-energy part of the kinetic-energy spectra, and would not have been included in the fission-fragment peak. It is thus assumed that the effect is real.

Such an effect could possibly arise if fission follows emission of heavy particles, say alpha particles, at about 90 deg to the beam. This would change the direction of the motion of the compound nucleus and yield angular distributions (for these cases) that would be peaked at angles away from 0 and 180 deg. However, it is not expected that heavy, high-energy particles are emitted frequently enough to explain the effect.

Although the angular distributions are shown as fitted by the Halpern and Strutinski theoretical curves, they have also been fitted to the distributions given by Griffin.⁹¹ The parameters that yielded the best agreement with the experimental data are listed in Table III.

Table III. Parameters and other values used in fitting angular distributions

System	P ($I_m^2/4K_O^2$)	r ($\bar{I}/\bar{K}$)	$(\bar{l})$	I_m^2	$E_{ex} - E_f$ (Mev)	
					H and S	Griffin
Au + 123.3-Mev C^{12}	10	5	45.2	4600	13.5	12
Au + 93.3-Mev C^{12}	7.2	4.25	33.5	2530	12	10
Au + 72.4-Mev C^{12}	6	4	22.3	1118	9	7
Au + 164.5-Mev O^{16}	11	~5.2	61.0	8380	36	15.5

In principle, one should determine the amount of fission that takes place at each stage in the chain of excited nuclei resulting from the evaporation of neutrons, protons, etc., and calculate the angular distribution for each stage. The over-all angular distribution would be obtained by weighting each distribution by its probability of occurrence and summing over all the fissioning species. However, the fission probabilities are not well known for the various nuclei involved here. Therefore the angular distributions are used to estimate the average excitation energy at which fission takes place, and from this result it is possible to determine some average fissioning species.

Because of barrier-penetration phenomena in compound-nucleus formation, Halpern and Strutinski note that the term I_m is not clearly defined, but may be approximated by $2 \bar{I}^2$, where $\bar{I}^2$ is the average value of the square of the spin of the compound nucleus. In keeping with that assumption,

$$2\bar{I}^2 = 2 \bar{l}^2 \quad (IV-6a)$$

$$\text{and} \quad 2\bar{I}^2 = \frac{9}{4} (\bar{l})^2 \quad (IV-6b)$$

are assumed here. These approximations are quite good for the high angular momenta involved in the reactions studied. The values of $\bar{l}$ listed in Table III were obtained from the compound-nucleus-formation data calculated by Thomas, using a square-well potential.²⁹ The $\bar{l}$ values

obtained by use of his parabolic-barrier model would be somewhat lower than the values used, but would not grossly change the conclusions drawn from the results. The assumption is also made that the spin of the compound nucleus is not changed by particle evaporation. This approximation has been justified by Halpern and Strutinski.

The values of $(E_{\text{ex}} - E_f)$ obtained by analysis of the data are listed in Table IV. The values obtained via the Halpern and Strutinski method were taken from their empirical curve of K_0^2 as a function of $(E_{\text{ex}} - E_f)$. Those following from the Griffin treatment were read from the linear curve of $\bar{K}$ vs $(E_{\text{ex}} - E_f)$. The values obtained by the two methods agree rather well except for the case of $\text{Au} + 164.5\text{-Mev } O^{16}$. In that case there is some question on which curve of Griffin's should be used.

In order to determine the average excitation energy at which fission occurs, one must estimate the fission barrier for the fissioning species. This is a difficult problem in that one does not know a priori which nucleus is fissioning. As noted below, there is considerable evidence for emission of charged particles as well as neutrons prior to fission. For nuclei in the At-Po region, Fairhall and Neuzil's results indicate fission barriers of the order of 18 Mev.⁹² W. J. Swiatecki has estimated a value of about 13.5-Mev for At^{206} .⁹³ This figure is based on the dependence of E_f on Z^2/A in the heavy element region, and the deviation of actual ground-state masses from the smooth mass surface calculated according to Green's equation.⁹⁴ This calculated value is probably more meaningful than the value obtained from analysis of the $\text{Bi}^{209} + \text{He}^4$ fission data cited above, because the fissioning nuclei studied in this work are very neutron-deficient compared with those investigated by Fairhall and Neuzil (Mostly At^{213}). For example, note that the value of Z^2/A for At^{206} (35.07) is approximately the same as that for Th^{230} (35.2).

There is another effect which complicates the choice of E_f . Pik-Pichak has shown that, owing to high angular momenta of the compound nuclei, there is a distortion from sphericity which increases the nuclear moment of inertia, thereby decreasing the rotational and Coulomb energies and increasing the surface energy.³⁰ The net effect is to lower the fission barrier relative to its height at low angular momenta. Estimates

Table IV. Properties of average fissioning species

System	$E_{ex} - E_f$ (H and S) (Mev)	$E_{est.} + E_f$ (Ref. 95) (Mev)	ΔE_f (Rotation) (Mev)	E_{ex} (H and S) (Mev)	No. evap. particles	Fissioning species
Au+123.3-Mev C ¹²	13.5	9.8	2.05	22.3	7n	At ²⁰²
		12.2	2.2	23.5	p5n	Po ²⁰³
		12.7	2.25	24.0	α 5n	Bi ²⁰⁰
		14.6	2.3	25.8	p α 4n	Pb ²⁰⁰
		15.8	2.4	26.9	2 α 3n	Tl ¹⁹⁸
Au+93.3-Mev C ¹²	12	11.0	1.1	21.9	4n	At ²⁰⁵
		12.9	1.3	23.6	p3n	Po ²⁰⁵
		14.1	1.3	24.8	α 2n	Bi ²⁰³
		16.1	1.4	26.7	p α n	Pb ²⁰³
		17.4	1.4	28.0	2 α	Tl ²⁰¹
Au+72.4-Mev C ¹²	9	11.6	0.5	20.1	3n	At ²⁰⁶
		13.9	0.6	22.3	pn	Po ²⁰⁷
		14.8	0.8	23.0	α n	Bi ²⁰⁴
Au+164.5-Mev O ¹⁶	36	8.3	3.0	41.5	7n	Fr ²⁰⁶
		9.7	3.2	42.6	p6n	Rn ²⁰⁶
		10.7	3.3	43.4	α 5n	At ²⁰⁴
		12.5	3.6	44.9	p α 4n	Po ²⁰⁴
		13.6	3.7	45.9	2 α 3n	Bi ²⁰²
		16.1	4.0	48.1	2 α p α n	Pb ²⁰³
		16.7	4.3	48.4	3 α 2n	Tl ²⁰⁰

of the magnitudes of this effect are listed in Table IV.⁹⁵ Since accurate mass data, necessary in the Swiatecki method of fission-barrier calculation, were not available for most of the nuclei in question, the Pik-Pichak calculations have also been used in calculating the fission barriers at zero angular momentum.

The rotational-energy effect also may require some correction to the expressions for angular distributions. This has been noted in a later paper by Pik-Pichak.⁹⁶ In all the angular-distribution calculations mentioned above, it has been assumed that the probability of fission is independent of I and K . However, because of the rotational-energy effect, Γ_f/Γ_n is expected to be an increasing function of I (for constant E_{ex}). Thus, in the angular-distribution expressions, the probability of a given I should be multiplied by a weighting function which describes this effect. Pik-Pichak has given the proper form for the corrected expression,⁹⁶ but there are not sufficient fission-probability data available to make this correction. The effect would cause the average I of the fissioning nuclei to be higher than the average I of the compound nuclei. This would mean that the $E_{ex} - E_f$ quantities inferred from the angular distributions are too low. Note that errors introduced by this effect would tend to cancel any errors in the assumption that no angular momentum is carried off by evaporation of particles prior to fission.

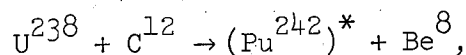
It is to be observed that the excitation energy at which fission takes place is nearly constant with bombarding energy, for $Au + C^{12}$. The excitation energies are rather low, indicating that fission takes place near the end of the chain of de-exciting nuclei, at least for the $Au + C^{12}$ cases. It is possible to estimate the numbers of various types of particles that could be emitted prior to fission. These estimates, listed in Table II, were obtained from the initial excitation energies and particle-binding energies (from Cameron's masses⁹⁷), with estimates of nuclear temperatures from the spallation data, and charged-particle energy spectra based on the equations of Dostrovsky, Fraenkel, and Friedlander.⁹⁸ In making the calculation, account has been taken of the lowered Coulomb barriers against charged-particle evaporation as found by Knox, et al.⁹⁹

More will be said concerning the implications of these results in the discussion of cross sections.

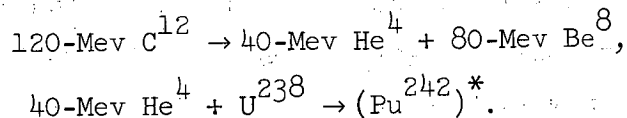
B. Momentum Transfer

The values obtained for η^2 (see Table II, Section II) from the various measured quantities indicates that, within the limits of error, fission of gold results from nuclear reactions in which the entire momentum of the bombarding particles is deposited in the fissioning system. This result has also been found by Alexander from fragment-recoil-range measurements.¹⁰⁰ This implies that "breakup" reactions,¹⁰¹ in which the bombarding particle breaks up, with only part of it penetrating the target nucleus (for example, $C^{12} \rightarrow Be^8 + He^4$, $He^4 + Au^{197} \rightarrow Tl^{201}$), do not result in many fission events in this system. Conversely, for the case of U^{238} bombarded with 120-Mev C^{12} ions, both Alexander¹⁰⁰ and Larsh et al.,¹⁰² find considerably less than full momentum transfer.

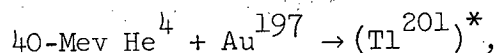
These results may be interpreted in the following manner: The fission barrier of plutonium isotopes is small enough that the fission probability is quite large for the compound nucleus resulting from the reaction



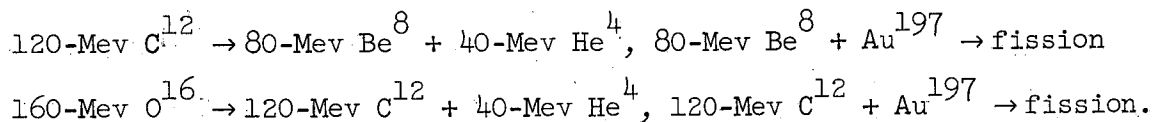
or the corresponding reaction in which the Be^8 is absorbed by the target nucleus. Note that in the example, one may consider the reaction as a two-stage process:



For the second reaction, Vandenbosch et al. report that the fission cross section is approximately equal to the cross section for formation of the compound nucleus.¹⁸ However, for the corresponding reaction in the bombardment of gold,



Fairhall and Neuzil have found $\sigma_f/\sigma_{\text{comp}}$ to be $\sim 10^{-4}$.⁹² Since the Au + 120-Mev C¹² fission cross section is large, one might expect fission from reactions of the type



The absence of evidence for such reactions is in agreement with the results of Ghiorso and Sikkeland, who find the probability for this type of breakup reaction to be smaller than for those in which only the alpha particle enters the nucleus.¹⁰¹

C. Kinetic Energy Release

It is apparent from the data shown in Fig. 23 of Section II that there is very little, if any, dependence of the most probable fragment kinetic energy upon energy of the bombarding particle. This result is in agreement with previous investigations on the effect of bombarding energy upon fragment kinetic energies.¹⁰³⁻¹⁰⁵ If fragment kinetic energies arise from Coulomb repulsion of the fragments after scission, (see Appendix B) these results indicate that the shapes of the fragments and the distance between their charge centers do not change with increasing excitation energy of the fissioning nucleus. The added energy presumably goes into evaporation of more prompt neutrons from the fragments.⁹ In the cases studied in this work, the results may be explained as being due to two effects. First, the Au + C¹² angular distribution experiments show that the excitation energy at which fission occurs increases only slightly (approx 2 Mev) when the bombarding energy is increased from 72.4 to 123.3 Mev. The slight increase in fragment kinetic energy indicated for the Au + C¹² case at higher bombarding energies results from emission of a larger average number of neutrons prior to fission than in the case of the lower energy bombardments. According to the Coulomb repulsion model mentioned above, the kinetic energy release should be proportional to $Z^2/A^{1/3}$ of the fissioning

nucleus. Thus the lower value of $A^{1/3}$ in the denominator would lead to a slightly higher kinetic-energy release for the higher bombarding energy.

Terrell has rather successfully correlated average total kinetic energy release with $Z^2/A^{1/3}$ for a series of heavy-element nuclides, (thorium and above), corroborating the Coulomb-repulsion model.²² However, if this model is strictly true, the total kinetic-energy release as a function of mass ratio - (mass of the heavy fragment)/(mass of the light fragment) - for a given fissioning nucleus should be a maximum for a mass ratio of unity, or symmetric fission. In all the time-of-flight measurements of kinetic energies of fragments from spontaneous and thermal-neutron fission in the heavy elements, there are indications for a dip in the total kinetic energy release at a mass ratio of unity.^{43,106,107} It was of interest to find out how the kinetic energies from the heavy-ion-induced fission would fit in with the data obtained at low energies.

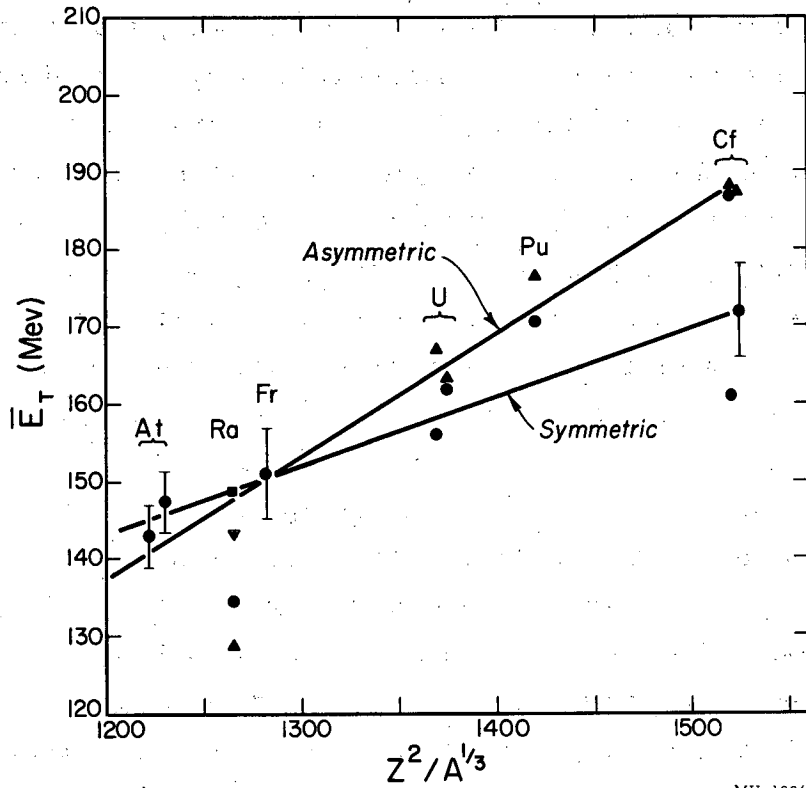
The data have been treated in a manner slightly different from Terrell's method in order to show up any possible connection between energy release and type of fission, symmetric or asymmetric. For the time-of-flight data listed in Table V, values for the total kinetic energy release for symmetric and most probable (asymmetric) modes have been taken from the graphs shown by the authors. Unfortunately, in all cases, the data do not extend into a mass ratio of unity (because of the small frequency of observation of those events), making it necessary to extrapolate the data. The values for neutron fission of Ra^{226} have been taken from the authors' kinetic energy spectra.⁴⁰ The point for $\text{U}^{238} + 123\text{-Mev C}^{12}$ was obtained from the experiments done with UF_4 targets (see Appendix A). The other heavy-ion-induced fission kinetic energies have been discussed above.

Table V. Total kinetic energy release in fission

Ref.	System	Average total K.E. release (Mev)		Assumed fission- ing nucleus	Counting method
		Symmetric	Asymmetric		
43	Cf ²⁵² Spont.fission	161	188	Cf ²⁵²	TF
106	Cf ²⁵² Spont.fission	187	187	Cf ²⁵²	TF
*	U ²³⁸ +123-Mev C ¹²	172±6	---	Cf ²⁵⁰	GS
107	Pu ²³⁹ +Thermal n	170.5	176.5	Pu ²⁴⁰	TF
107	U ²³⁵ + " "	156	167	U ²³⁶	TF
107	U ²³³ + " "	162	163	U ²³⁴	TF
40	Ra ²²⁶ +14.7-Mev n	134.5	---	Ra ²²⁷	GS
40	Ra ²²⁶ +4-Mev n	---	128.5	Ra ²²⁷	GS
*	Au ¹⁹⁷ +164.5-Mev O ¹⁶	151±6	---	Fr ²⁰⁶	GS
*	Au ¹⁹⁷ +123-Mev C ¹²	147.5±4	---	At ²⁰³	GS
*	Au ¹⁹⁷ +72.4-Mev C ¹²	143±4	---	At ²⁰⁶	GS

*-This work. TF- Time-of-flight. GS- Gas Scintillation.

The fragment kinetic energies from the counting experiments have been corrected for emission of prompt neutrons. The average number of prompt neutrons, $\bar{\nu}$, that would be emitted in spontaneous fission of the nuclei in question were estimated from the curves shown in Fig. 8 of a paper by Bondarenko et al.¹⁰⁸ The increase in $\bar{\nu}$ with excitation energy was also obtained from data contained in their paper. Excitation energies at the time of fission and the fissioning nuclei in the gold experiments were determined from the results of the angular distribution experiments. For U²³⁸ + C¹², the fissionability of Cf²⁵⁰ is so large that it has been assumed that fission takes place in the first stage of the chain of de-exciting nuclei. In accord with Terrell's results, 0.8 Mev has been added to the total kinetic energy release for each prompt neutron. The resulting kinetic energies are listed in Table V and are plotted as a function of $Z^2/A^{1/3}$ (of the assumed fissioning nucleus) in Fig. 37.



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Fig. 37. Average total kinetic energy release in fission as a function of $Z^2/A^{1/3}$ of the fissioning nucleus.

- - Symmetric events
- ▲ - Asymmetric events.
- - "Corrected" symmetric events, Ra^{227}
- ▼ - "Corrected" asymmetric events, Ra^{227} .

The most obvious conclusion to be drawn from the results presented in Fig. 37 is that the data, especially for symmetric fission, are in poor shape. There is very poor agreement between the two symmetric-fission points obtained for Cr^{252} by the two groups of experimenters. However, those points do bracket the Cr^{250} point obtained in this work. One could argue that, in $\text{U}^{238} + \text{C}^{12}$ fission, the average fissioning nucleus is somewhat lower in Z than californium, due to direct-interaction reactions ("breakup," see above), induced fission, or evaporation of charged particles prior to fission. However, it seems quite unlikely that such processes occur frequently enough to represent the most probable fragment kinetic energy. It might also be argued that the average fissioning nuclei in the gold experiments are lower in Z than has been assumed. This point is not unreasonable on the basis of the other results of this study, but it would not change the main features of the results. The points plotted for fission of Ra^{227} do not agree at all with the other results. On the assumption that this is due to an experimental error, rather than an anomaly, the two Ra^{227} points have been raised by an amount that places the symmetric-fission point on the line determined by the other symmetric-fission points.

In view of the crossover of the two curves in the vicinity of actinium, it is tempting to correlate kinetic energy release with relative probability for symmetric and asymmetric fission events, which also has a crossover in this region for low-energy fission.¹⁶ If such a correlation does exist, it is not in agreement with the statistical theory of Fong,¹⁴ which was later extended by Cameron.¹⁵ According to that model, the probability of a given mode of fission is proportional to the density of final states to which that type of division leads. This contains a term proportional to the kinetic-energy release and two exponential terms for the level densities of the fragments as functions of their excitation energies. Although a larger kinetic energy release would increase the first term mentioned, it would decrease the amount of excitation energy available to the fragments, thus decreasing the two exponential factors. One would expect the decrease in the values of the

exponential terms to override the increase in the term that is proportional to energy release.

Swiatecki and Alexander have found rather convincing evidence that, for Cf^{252} spontaneous fission, the dip in kinetic energy release at a mass ratio of unity may be due to a high probability for emission of long-range alpha particles in symmetric-fission events.¹⁰⁹ If this is true, one could interpret the results presented in Fig. 37 as indicating that the probability for emission of long-range alpha particles associated with symmetric-fission events is an increasing function of Z , or $Z^2/A^{1/3}$.

One result that is difficult to reconcile with the ideas presented is the observation by Douthett and Templeton that the dip in kinetic-energy release in symmetric fission seen at low excitation energies in the uranium region is not present in the fission of U^{238} with 18-Mev deuterons.¹⁰⁴ This result, based on the recoil range of one fission product, should probably be investigated in more detail.

It is obvious that before one can draw many conclusions concerning the ideas advanced here, many more and more accurate results must be obtained on dependence of total kinetic energy release on mass ratio, excitation energy, and Z and A of the fissioning nucleus. Data are particularly needed for symmetric fission in the actinide elements. It would be useful also to have information from fission at excitation energies intermediate between thermal-neutron- and heavy-ion-induced fission.

The results obtained on kinetic energy release may be summarized:

(a) The kinetic energy release resulting from heavy-ion-induced fission appears to be approximately the same as for symmetric fission in neighboring nuclei at lower excitation energies. There is no apparent effect on this quantity due to the higher excitation energies and angular momenta involved in heavy-ion reactions.

(b) There is evidence that the total kinetic energy release is greater for asymmetric fission than for symmetric fission of elements above the radium-actinium region. The converse is apparently true below this region.

D. Cross Sections

Spallation Reactions

Previously, excitation functions for neutron-evaporation reactions in the heavy-element region have been interpreted on the basis of a model proposed by Jackson¹⁹ and modified to include the effects of fission by Vandenbosch et al.¹⁸ Although, as noted below, the assumptions inherent in the model are undoubtedly poor when applied to heavy-ion-induced nuclear reactions, it is the only model available by which cross sections may be predicted by simple, analytical methods. This is the justification for its use.

The following assumptions were made in the original derivation of the expression for neutron-evaporation cross sections:¹⁹

(a) The energy spectrum of neutrons emitted by a compound nucleus of excitation energy E , and nuclear temperature T (of the residual nucleus), is of the form $\epsilon \exp(-\epsilon/T)$, where ϵ is the kinetic energy of the neutron.

(b) Neutron emission occurs whenever it is energetically possible.

(c) Proton evaporation is neglected.

(d) The nuclear temperature is constant with increasing excitation energy.

As a result of these assumptions, the probability $P(x,E)$ for emission of exactly x neutrons from a compound nucleus originally at an excitation energy E is given by

$$P(x,E) = I(\Delta_x, 2x - 3) - I(\Delta_{x+1}, 2x - 1), \quad (\text{IV-7})$$

where $I(x,n)$, is Pearson's incomplete gamma function,

$$I(z,n) = \left(\frac{1}{n!}\right) \int_0^z x^n e^{-x} dx, \quad (\text{IV-8})$$

and

$$\Delta_x = \frac{E - \sum_{i=1}^x B_i}{T}, \quad (\text{IV-9})$$

and B_1 is the binding energy of the i th neutron. It should be noted that $I(\Delta_x, 2x - 3)$ represents the probability for emission of at least x neutrons and $I(\Delta_{x+1}, 2x - 1)$ represents that for $x+1$ neutrons, the difference between the two terms being equal to the probability for emission of exactly x neutrons.

The cross section for evaporation of x neutrons is obtained by multiplication of $P(x, E)$ by the cross section for compound-nucleus formation, σ_{comp} . If other processes compete with neutron evaporation it is necessary to multiply the calculated cross sections by the product of the values of Γ_n/Γ_T (ratio of level width for neutron emission to total level width) for each of the x compound nuclei preceding the final product. When this modification is made, the expression for neutron-emission cross sections becomes

$$\sigma_{xn} = \sigma_{\text{comp}} \left(\frac{\Gamma_n}{\Gamma_T} \right)_1 \left(\frac{\Gamma_n}{\Gamma_T} \right)_2 \dots \left(\frac{\Gamma_n}{\Gamma_T} \right)_x [I(\Delta_x, 2x-3) - I(\Delta_{x+1}, 2x-3)]. \quad (\text{IV-10})$$

An attempt has been made to fit the experimental cross-section data from the $\text{Pt}(\text{N}^{14}, xn)\text{At}$ reactions with cross sections calculated according to Eq. (IV-10). There are several factors that make the calculations somewhat uncertain. One of these involves the selection of the proper values for σ_{comp} . Especially at bombarding energies in the region of the Coulomb barrier height, σ_{comp} depends quite sensitively upon the model chosen. The values used here are based on the calculations by Thomas for the square-well nuclear potential.²⁹ It must be observed that the sharp rise in the cross sections at low energies causes small errors in the energy of the bombarding particle to yield large errors in the magnitude of the measured cross sections. Also, there is considerable uncertainty in the energetics of the reactions. Where possible, the excellent mass and neutron-binding-energy data of Foreman and Seaborg¹¹⁰ have been used. These data, however, do not extend to astatine isotopes below mass 209 or to the target nuclei. Thus it has been necessary to obtain some of these values from the data of Wapstra¹¹¹ and Cameron.⁹⁷ By comparison with Foreman and Seaborg, it appears that

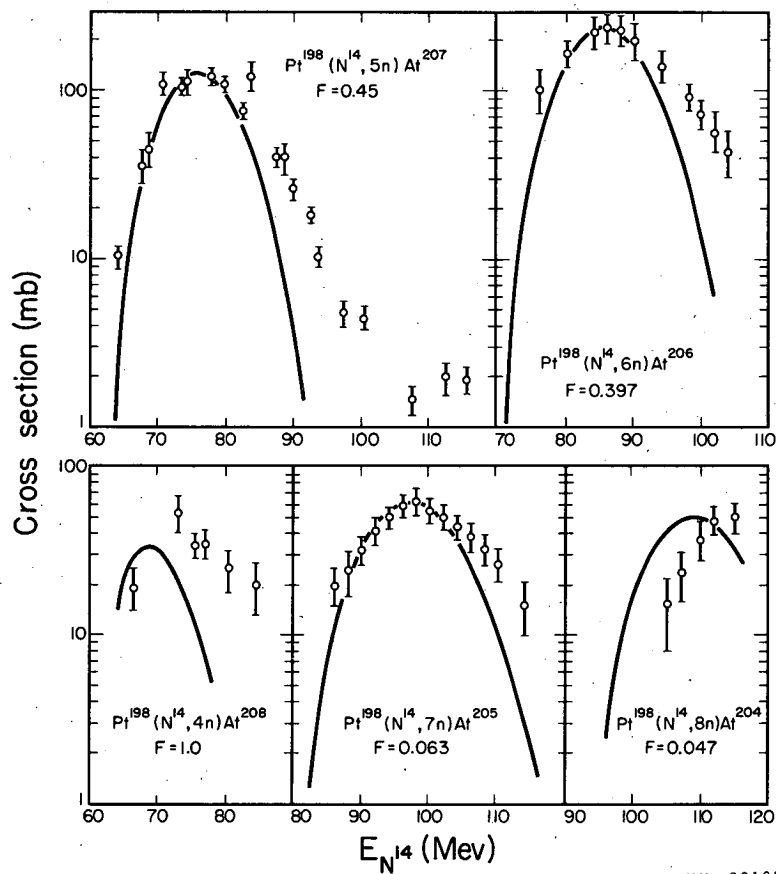
Cameron's calculated neutron binding energies are too high. For this reason, the calculated binding energies have been reduced to bring them into better agreement with the data of Foreman and Seaborg. Even so, the uncertainties in the individual neutron binding energies may be as much as 0.5 Mev. This is especially true for the lighter isotopes.

The comparison between calculated and experimental excitation functions are shown in Figs. 38 and 39. In fitting the magnitudes of the excitation functions, the product of level width ratios of Eq. (IV-10) has been given as the factor F .

The best fit to the experimental results was obtained with a nuclear temperature of 1.3 Mev. This is to be compared with values of about 1.35 Mev, which have been used in similar treatments of data from helium-ion-induced reactions in the actinide elements,^{5,18} and a value of 0.96 Mev, which was obtained by Ghiorso and Sikkeland from a study of heavy-ion-induced neutron-evaporation reactions in the heavy-element region.¹⁰¹ The meaning of the value of T used in fitting the experimental cross sections is not clear. No direct correlation of temperature with target projectile, or energy is apparent. Perhaps it is best to regard it simply as a parameter which may be adjusted to fit the position and shape of the excitation functions.

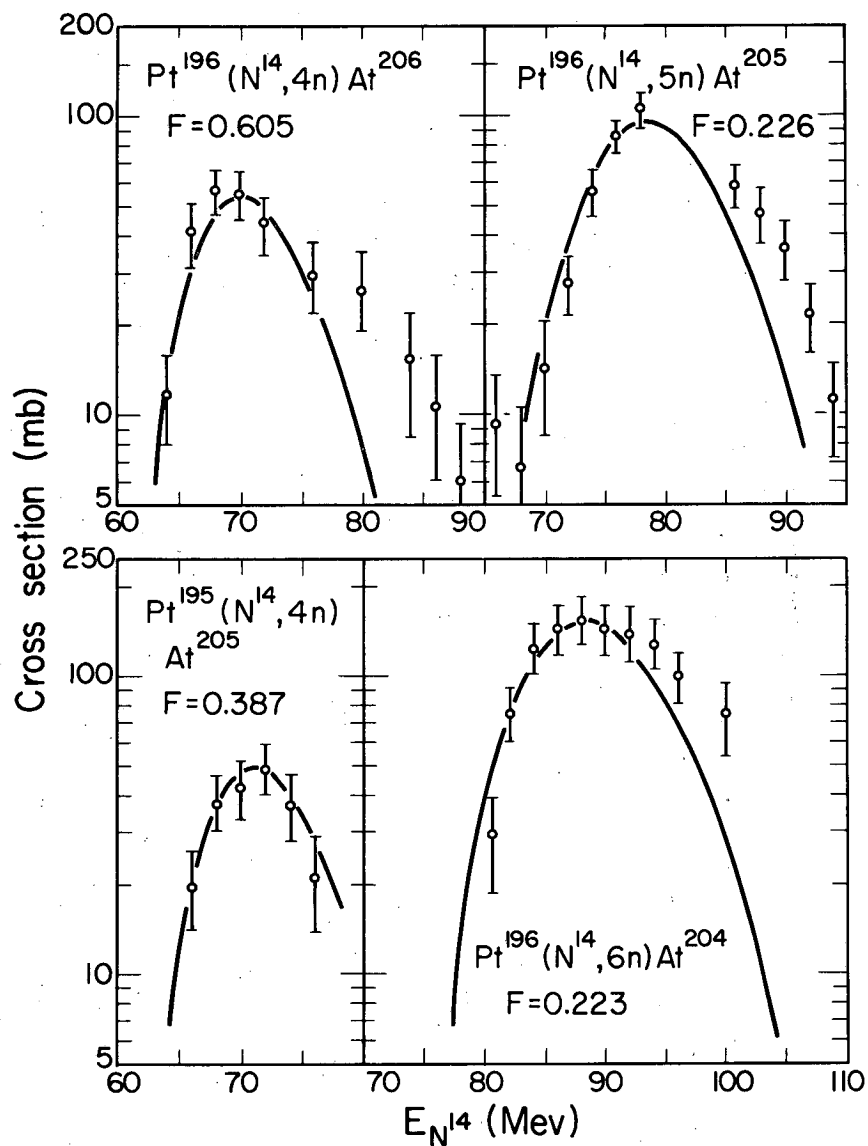
Explanations have been given for the apparent constancy of the nuclear temperature. According to one theory, increased excitation energy is used to break up ordered nuclear structure.¹¹² This may be compared to the melting of ice. This process continues up to an energy at which most of the ordered structure ("ice") has been broken up ("melted") before further increase in energy raises the nuclear temperature.

The agreement between the calculated shapes and experimental excitation functions on the high-energy side of the peaks is poor. This is probably because the Jackson treatment neglects the effects of the large angular momenta involved in heavy-ion-induced reactions. For example, the average angular momentum of the compound nuclei formed by bombardment of Pt^{198} with 100-Mev N^{14} ions is about 35 $\hbar$. The emission of the first few neutrons scarcely changes the average spin value, since



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Fig. 38. Excitation functions for the reactions $\text{Pt}^{198}(\text{N}^{14}, x\text{n})\text{At}^{212-x}$ for $x = 4, 5, \dots, 8$. Solid curves represent the excitation functions calculated according to the Jackson model with $T = 1.3$ MeV. $F = [(\Gamma_n/\Gamma_T)_1 (\Gamma_n/\Gamma_T)_2 \dots (\Gamma_n/\Gamma_T)_i]$.



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Fig. 39. Excitation functions for the neutron-evaporation reactions of Pt^{195} and Pt^{196} bombarded with N^{14} ions. Solid curves represent the excitation functions calculated according to the Jackson model with $T = 1.3$ MeV. $F = [(\Gamma_n/\Gamma_T)_1 (\Gamma_n/\Gamma_T)_2 \dots (\Gamma_n/\Gamma_T)_i]$.

at high excitation energies high-spin levels in the residual nuclei are plentiful^{31,112,113} (see Appendix B). Also, the transmission coefficient for passage of low-energy neutrons through the nuclear surface is a sharply decreasing function of the orbital angular momentum carried off by the neutron.¹¹⁴ Thus the main effect of early neutron-evaporation events is to broaden the distribution of spin values without appreciably lowering the average value. It is therefore quite possible for de-excitation to take place down to an excitation energy only slightly above the binding energy of the next neutron, while the spin value remains quite high.

Consider the fate of a nucleus which has an excitation energy only slightly - say, 2 Mev - above the binding energy of the next neutron, and which possesses a large amount of angular momentum. In order for it to emit another neutron, one or a combination of two things must occur. Either the decay must proceed to a high spin state at a low excitation energy of the residual nucleus, (≤ 2 Mev in this example) or the neutron must carry off a large amount of orbital angular momentum. Since the probability of occurrence of high spin levels in nuclei at low excitation energies is small, the first alternative is hindered. The second process is hindered for the reason given above. Owing to these effects, the probability for neutron emission may be decreased relative to other decay modes - such as a cascade of gamma rays, which lowers the excitation energy below the neutron binding energy.

According to the second assumption made in the Jackson treatment, neutron emission occurs whenever energetically possible. This is probably a very good approximation for low-angular-momentum reactions. However, because of the increased relative probability for gamma de-excitation from low-energy, high-spin states, many nuclei which would, according to the model, emit x neutrons may instead de-excite by gamma emission following evaporation of $x-1$ neutrons. On this basis one can explain the experimentally observed high-energy tails of the xn reaction excitation functions.

If the fission barrier is nearly equal to or less than the binding energy of the next neutron, instead of de-exciting by gamma emission, the nuclei in high-spin states may undergo fission. Fission is a process

that can easily take place from high spin levels. Even without using spin density arguments, Pik-Pichak has predicted increases in Γ_f/Γ_n with increasing angular momentum.³⁰ Thus when fission can occur at low excitation energies it may remove most of the nuclei that have high spins. In this case, one would expect the high-energy tails of the peaks in the neutron-evaporation excitation functions to be in better agreement with the Jackson calculations. The few cross-section data available for heavy-ion-induced reactions are in agreement with this picture. Ghiorso and Sikkeland's experimental xn-reaction cross sections from carbon bombardment of U^{238} and Pu^{242} are in rather good agreement with the shapes of the calculated excitation functions.¹⁰¹ The fission barriers for the californium and fermium compound nuclei are less than the neutron binding energies. On the other hand, the excitation functions for the (C^{12}, xn) and (N^{14}, xn) reactions of V^{51} have extensive high-energy tails, and the peaks occur at higher energies than for the corresponding reactions induced by protons.^{34,115} Fission is not a competing process in these reactions. As one would expect, the $Pt + N^{14}$ excitation functions are intermediate between these extreme cases.

On the basis of these arguments, the behavior of the high-energy tails of heavy-ion-induced neutron-evaporation reaction excitation functions may serve as a crude measure of the fission barrier. Detailed calculations would be necessary, however, before this concept could become very useful for estimating fission barrier heights.

In previous work, Γ_f/Γ_T has been assumed constant and equal to $\Gamma_f/(\Gamma_f + \Gamma_n)$ for a given nucleus.^{5,17,18} Thus analysis of spallation reaction data has yielded Γ_f/Γ_n values for the nuclei involved. Such a treatment has not been attempted in the present study. As noted below, there is evidence that Γ_f/Γ_n is dependent upon energy in the astatine compound nuclei. Also, it appears that one cannot neglect charged-particle emission from these compound systems, particularly at high excitation energies. Some considerations on the relative fissionabilities of the nuclei involved are given below.

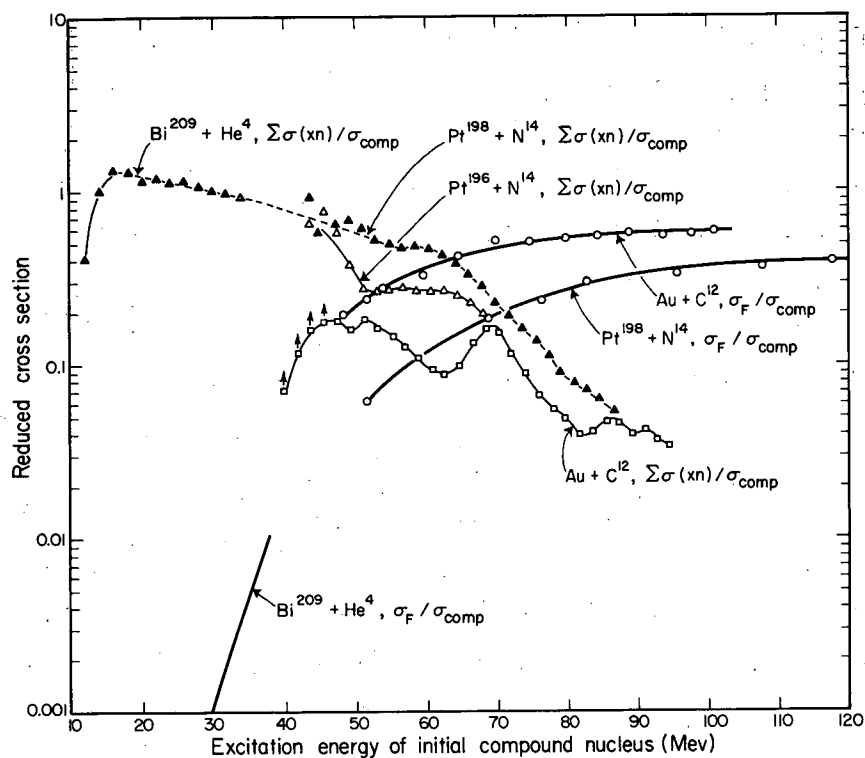
It should be noted that the agreement between calculated and experimental cross sections for the $4n$ and $8n$ reactions of $Pt^{198} + N^{14}$ is poor.

In the former, the disagreement likely arises from errors in σ_{comp} in that energy region. In the other case, the difficulty is probably experimental. As noted above, At^{204} is produced by several reactions involving the various platinum isotopes. This makes it difficult to obtain the $\text{Pt}^{198}(\text{N}^{14}, 8n)\text{At}^{204}$ cross sections from the data.

Fission and Spallation Probabilities

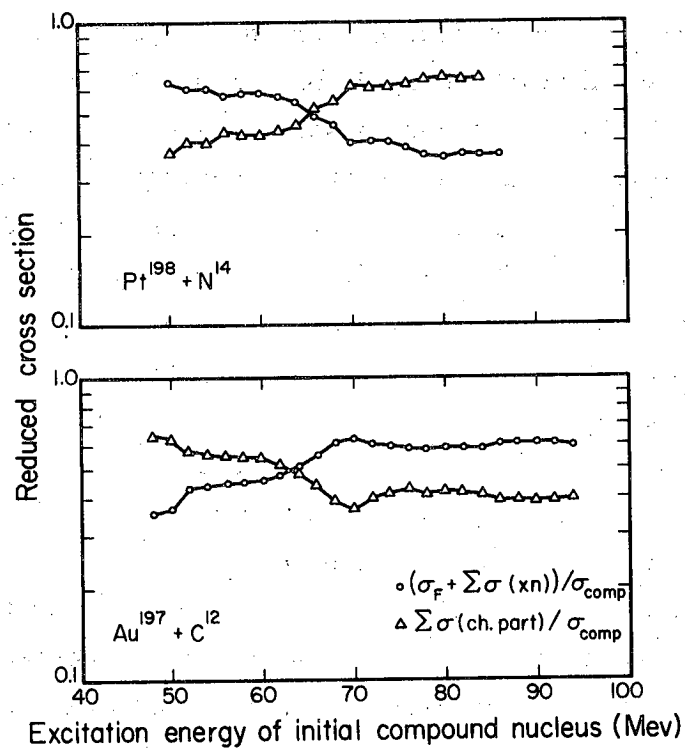
The cross-section data of this and other studies are summarized in Figs. 40 and 41. In order to obtain the probabilities for various modes of disintegration of the compound nuclei, all the cross sections have been divided by appropriate cross sections for compound-nucleus formation. The xn-reaction cross-section data for $\text{Au} + \text{C}^{12}$ were obtained from Latimer and Thomas.³⁵ It should be noted that at low energies the values shown for that system are lower limits, as the $(\text{C}^{12}, 3n)$ reaction has not been included in the summation. The $\text{Bi}^{209} + \text{He}^4$ spallation data were taken from the reports of Kelly and Segrè³⁶ and Vandenbosch and Huizenga.⁵

Fission cross sections for the $\text{Bi}^{209} + \text{He}^4$ system were obtained from Fairhall and Neuzil's results.⁹² In order to get some idea of the agreement between the fission cross sections determined in this study and those of other experimenters, a similar curve was constructed from the data of Polikanov and Druin for fission of gold with nitrogen ions.³³ The agreement is not as good as one would desire. The latter results are uniformly higher and rise more steeply with energy than for fission of gold with carbon ions. The first discrepancy noted could be explained as arising due to greater fissionability of the compound nuclei studied, because of the higher value of Z. Most of Polikanov and Druin's results were obtained by counting the fragments emitted in the forward hemisphere ($\theta_{\text{lab}} \leq 90$ deg) in a double ionization chamber. It is not clear that they have made corrections for center-of-mass motion. Comparison of all their results suggests that they have not. Their fission cross sections for nitrogen bombardment of bismuth appear to be larger than most reasonable estimates for compound-nucleus formation. Also they are larger than the corresponding cross sections for uranium. Since, as has been noted above, complete momentum transfer is not obtained in heavy-ion-induced fission of uranium, this result would be expected if corrections were not applied



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Fig. 40. Reduced cross sections for fission ($\sigma_F/\sigma_{\text{comp}}$) and neutron-evaporation reactions ($\Sigma\sigma(xn)/\sigma_{\text{comp}}$) as a function of excitation energy of the initial compound nucleus for various astatine compound nuclei.



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Fig. 41. Reduced cross sections, $\Sigma\sigma(ch. part.)/\sigma_{comp}$ and $[(\sigma_F + \Sigma\sigma_{xn})/\sigma_{comp}]$ as functions of excitation energy of the initial compound nucleus. Upper curves: $Pt^{198} + N^{14}$; lower curves: $Au^{197} + C^{12}$.

for center-of-mass motion. The correction would become more serious with increasing bombarding energy. The full-energy $C^{12} + Au$ fission cross section obtained in the present work is in reasonable agreement with a value of 900 mb reported by Blann from radiochemical studies of the fission products.¹¹⁶

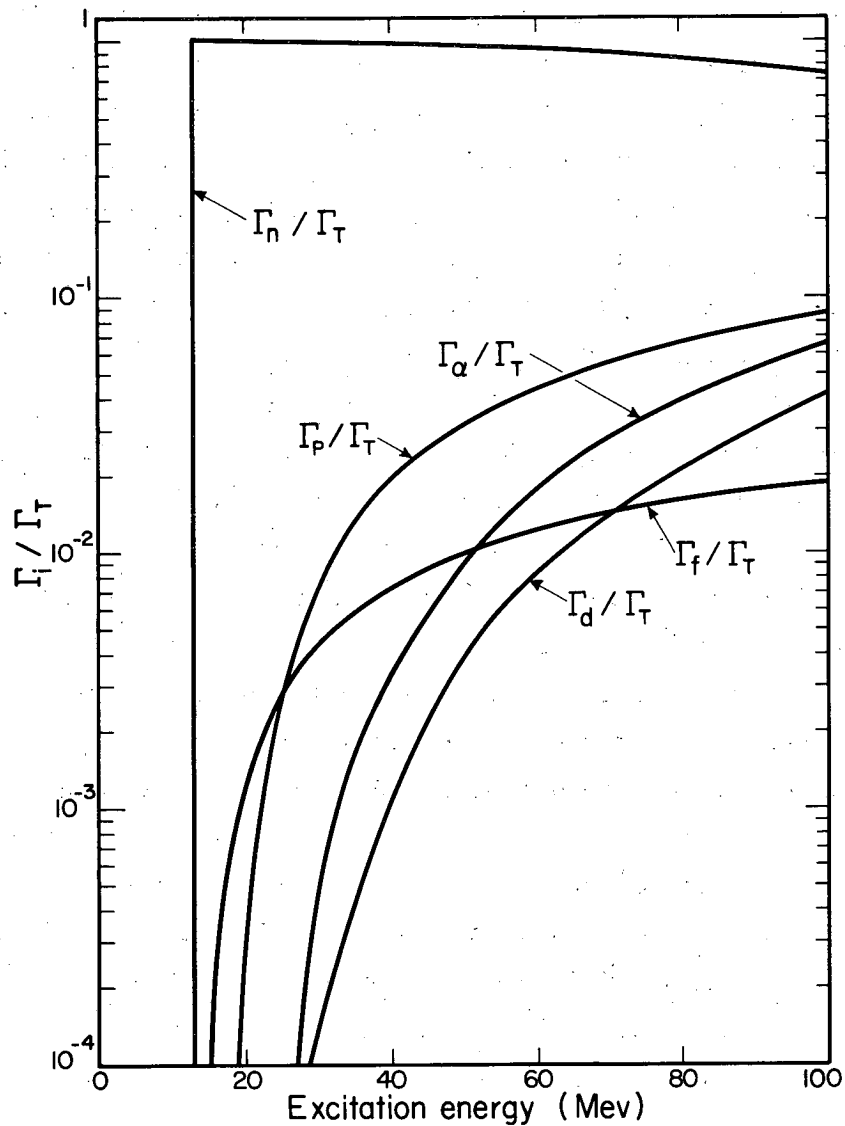
The curves given in Fig. 41 for $\Sigma\sigma(\text{charged particles})/\sigma_{\text{comp}}$ represent those processes in which charged particles were emitted without being followed by fission. The curves were obtained by simply subtracting the fission and neutron-evaporation cross sections from the cross section for compound-nucleus formation. Admittedly, this analysis is subject to large possible uncertainties. Perhaps the part most susceptible to error is the choice of σ_{comp} . To be consistent, all the compound-nucleus-formation cross sections are based on a square-well nuclear potential with a radius parameter of 1.5×10^{-13} cm.^{29,114} The reduced spallation cross sections at energies near the Coulomb barrier show rapid fluctuations, suggesting errors in σ_{comp} or the experimental data or both. Also, at the highest energies, it has not been shown that the calculated compound-nucleus-formation cross sections are correct. Owing to rotational-energy requirements and possible "contact-transfer" processes, the probability for formation of compound nuclei with extremely high spin values may be reduced below the calculated values.^{99,117} Errors in the charged-particle cross sections may also result from combinations of errors in the fission and neutron-evaporation cross sections.

Several observations may be made from the results shown in Figs. 40 and 41:

(a) Fissionability is an increasing function of Z^2/A . The fission probability for Pt^{198} bombarded with nitrogen ions (initial compound nucleus, At^{212}) is less than that for $Au^{197} + C^{12} (\rightarrow At^{209})$. Also the neutron-evaporation cross sections decrease in proceeding from the initial compound nucleus At^{212} to At^{210} ($Pt^{196} + N^{14}$) to At^{209} . (Note that the latter results may be partly due to increased probability for charged-particle emission from the more neutron-deficient isotopes because of their higher neutron binding energies and lower charged-particle binding

energies.) That fissionability increases with decreasing A for a given Z is in disagreement with the statement made by Fairhall, Jensen, and Neuzil,¹⁶ but in agreement with their later experimental results on fission of the various lead isotopes by helium-ion bombardment (with the exception of Pb^{206} , which does not follow the trend of the other isotopes).⁹²

(b) Some fission must occur following charged-particle emission from the compound nuclei formed by carbon bombardment of gold. This may also happen with $\text{Pt}^{198} + \text{N}^{14}$, especially at the highest energies, but in this case the evidence is not as clear-cut. This result is at least in qualitative agreement with the mass-yield data of Blann¹¹⁶ and the fission and charged-particle-evaporation data of Knox et al. in the system $\text{Au}^{197} + 160\text{-Mev } \text{O}^{16}$.⁹⁹ Justification for this observation is based on calculations done according to the model used by Doestrovsky, Fraenkel, and Friedlander (hereafter referred to as DFF).⁹⁸ Results of the calculations are shown in Fig. 42. The curves shown represent the probability for de-excitation by the various decay modes as a function of excitation energy for the At^{209} compound nucleus formed by $\text{Au}^{197} + \text{C}^{12}$. In performance of these calculations some modifications of the DFF method have been made. As has been suggested by Ericson and Strutinski, collective rotational energy has been subtracted from the total excitation energy before calculation of nuclear temperatures appropriate to the evaporation processes.¹¹³ Fission branching ratios were calculated by substitution of fission barrier heights determined from Pik-Pichak's equations³⁰ (as corrected by J. Hiskes⁹⁵) into the equation for Γ_f/Γ_n given by Doestrovsky, Fraenkel, and Rabinowitz.¹¹⁸ In all cases, the calculations refer to compound nuclei formed with the average values of I_{comp}^2 , obtained from Thomas' calculations.²⁹ The resulting curves indicate that the probability for emission of various charged particles increases rapidly with excitation energy. The absolute calculated values for charged-particle emission are probably much too low. Knox et al. find for $160\text{-Mev } \text{O}^{16} + \text{Au}^{197}$ that the charged-particle evaporation spectra are displaced to considerably lower energies than predicted by calculations based on the DFF model.⁹⁹ Such an effect would greatly increase the charged-particle emission probabilities. Compare now the curve of $\Sigma\sigma(\text{ch. part.})/\sigma_{\text{comp}}$ for $\text{Au} + \text{C}^{12}$



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Fig. 42. Calculated probabilities (Γ_i/Γ_T) for various modes of de-excitation of the compound nucleus At^{209} as a function of excitation energy.

with the curves for $\Gamma_{\text{ch. part.}}/\Gamma_T$. That the experimental curve for $\Sigma\sigma(\text{ch. part.})/\sigma_{\text{comp}}$ does not increase rapidly with energy suggests that many of the charged-particle emissions are followed by fission events. Similar arguments suggest that this effect occurs in the $\text{Pt}^{198} + \text{N}^{14}$ system at the higher energies, but not with as large a probability as in the case of $\text{Au}^{197} + \text{C}^{12}$.

(c) As predicted by Fairhall et al., Γ_f/Γ_T appears to increase with increasing excitation energy up to about 50 Mev.¹⁶ Beyond that energy region, there is evidence that it decreases. These observations are based on the variations of $\sigma_f/\sigma_{\text{comp}}$ with excitation energy of the initial compound nucleus. Although there is a fairly wide region of energies in which there are no data points, the curve obtained for fission of Pt^{198} with nitrogen ions appears to be an extension of the results for $\text{Bi}^{209} + \text{He}^4$ (compound nucleus, At^{213}). As the excitation energy is increased, it is energetically possible to evaporate more particles, thus there are more chances for fission to compete with other decay processes. Even if Γ_f/Γ_T stayed constant with increasing excitation energy $\sigma_f/\sigma_{\text{comp}}$ would continue to rise. The decrease in Γ_f/Γ_T at higher excitation energy is probably due mainly to increased competition from charged-particle evaporation early in the de-excitation process. Also, Halpern has shown that, on theoretical grounds, one would expect a decrease in Γ_f/Γ_n beyond some high excitation energy.⁸ However, in view of the fairly large fission barriers in the astatine region compared with the neutron binding energies, it is doubtful that the effect which he discussed is important in this case. Level-density and angular-momentum arguments that suggest an anomalously large value of Γ_f/Γ_n at very low excitation energies are discussed below.

E. Summary

In this section it is desirable to examine the results of the angular-distribution experiments in the light of the ideas advanced in the preceding section. Recall that the results indicate that for $\text{Au} + \text{C}^{12}$, fission occurs at an average excitation energy of about 20 to 25 Mev, nearly independent of bombarding energy. I believe that the following picture explains, at least qualitatively, all the results found in this study.

First, charged-particle emission is very important. The probability for emission of charged particles from the initial compound nucleus increases rapidly with excitation energy. This decreases the relative probability for fission in the initial compound nucleus. Charged-particle emission decreases the value of Z^2/A . Following either neutron or charged-particle emission from early stages of the de-excitation process, there are two trends that would predict additional neutron emission prior to fission. On the one hand, fissionability appears, from the results given herein and from those of previous investigations, to increase with increasing Z^2/A .^{18,20,92,119} Secondly, there are the angular-momentum and level-density arguments which were described above in the discussion of the shapes of excitation functions for neutron-evaporation reactions. The average angular momentum of the initial compound nuclei increases with increasing bombarding energy. Early evaporation of particles has very little effect on the distribution of spin values.* Thus, in many cases, de-excitation proceeds to a nucleus that has an excitation energy of the order of 15 Mev and a large spin value. At these low excitation energies, charged-particle emission is very unlikely. Neutron emission is greatly hindered because of the low probability for high-spin states in the residual nucleus (see Appendix B). Thus fission, a process in which large amounts of angular momentum may be removed in the form of

* Preliminary calculations by T. D. Thomas¹²⁰ based on the equations of Ericson and Strutinski^{112,113} indicate that evaporation of one neutron from At^{209} at 95 Mev of excitation energy and spin of 66.5 $\hbar$ decreases the spin by approximately 1 unit ($\hbar$).

fragment spins and mutual orbital angular momentum, may become quite predominant. The unusually large fission probability at low excitation energies would be expected to increase with increasing bombarding energy. The ratio $\sigma_f/\sigma_{\text{comp}}$ does not increase at higher bombarding energies because fewer nuclei with the Z of the original compound nucleus survive the early evaporation stages. The increasing number of lower-Z nuclei have lower fission probabilities. At excitation energies on the order of the fission barrier and lower, the probability for de-excitation by gamma-ray cascades may be much higher than expected (again because of hindrance of neutron emission), giving rise to large high-energy tails on the excitation functions for neutron-evaporation reactions.

It is unfortunate that a simple quantitative investigation of these notions is not possible. Probably the most feasible check on the quantitative agreement between the experimental data and the ideas advanced would be obtained via a Monte Carlo method. It should be noted that, although they are not directly applicable to this work, the Monte Carlo spallation-fission competition calculations of Doestrovsky, Fraenkel, and Rabinowitz are in rather good qualitative agreement with the experimental results.¹¹⁸ In particular, their calculations predict the occurrence of fission following evaporation of small particles.

Before any complete understanding of the mechanisms of heavy-ion-induced fission can be approached, there is a need for more experimental data in several specific areas. In particular, cross sections for production of nuclei resulting from charged-particle evaporation are definitely needed. As in most regions, these data would be very difficult to obtain in the astatine region because of the short electron-capture and alpha-decay half lives of the xn-evaporation products. Also, it would be desirable to have information about total reaction cross sections in order to obtain a better idea of the dependence of cross section for compound-nucleus formation upon bombarding energy.

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I wish to thank my wife, Connie, for her patience in the face of long and erratic bombardment schedules, her encouragement and her assistance with the grammatical construction of this thesis.

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"This is the way the world ends
Not with a bang, but a whimper."

T. S. E.

APPENDICES

APPENDIX A. Supplementary Fission-Counting Experiments

1. Faraday Cup Experiments

Method

A considerable amount of time was spent investigating the properties of Faraday cups. In order to obtain accurate absolute cross sections it was necessary to determine the validity of the beam-current readings obtained by use of the Faraday cup.

There are three major possible sources of error:

(a) Pickup of electrons. When the heavy-ion beam passes through absorber or target foils, many electrons are ejected from the foil. If these electrons are picked up by the cup, their charges cancel some of the positive charge of the beam particles. This effect yields beam-current readings that are too low.

(b) Loss of secondary electrons. When the heavy-ion beam strikes the Faraday cup many electrons may be ionized and released at the surface of the metal. If these electrons escape from the cup, a higher positive current is indicated, as loss of negative charges is equivalent to acceptance of positive charge.

(c) Loss of beam particles. In order to obtain absolute cross sections, one should measure the number of particles that strike the target. The number of particles absorbed by nuclear reactions is small in the thin targets used for these experiments. However, many of the beam particles are elastically scattered by the target nuclei. The beam-current readings are too low if the Faraday cup does not subtend a large enough angle to accept most of these scattered particles. Fortunately, the differential scattering cross section decreases very sharply with increasing angle (approximately proportional to $1/\sin^4(\theta/2)$ for pure Rutherford scattering¹²¹).

The first two effects may be eliminated by placing the Faraday cup in a magnetic field whose lines of force are perpendicular to the beam direction. This curves the paths of electrons attempting to enter or

leave the cup. Alternatively, these effects may be removed by applying a negative potential to a grid placed near the entrance to the cup. This repels electrons that would enter or leave the cup. The third effect may be minimized by using a cup that subtends a large solid angle.

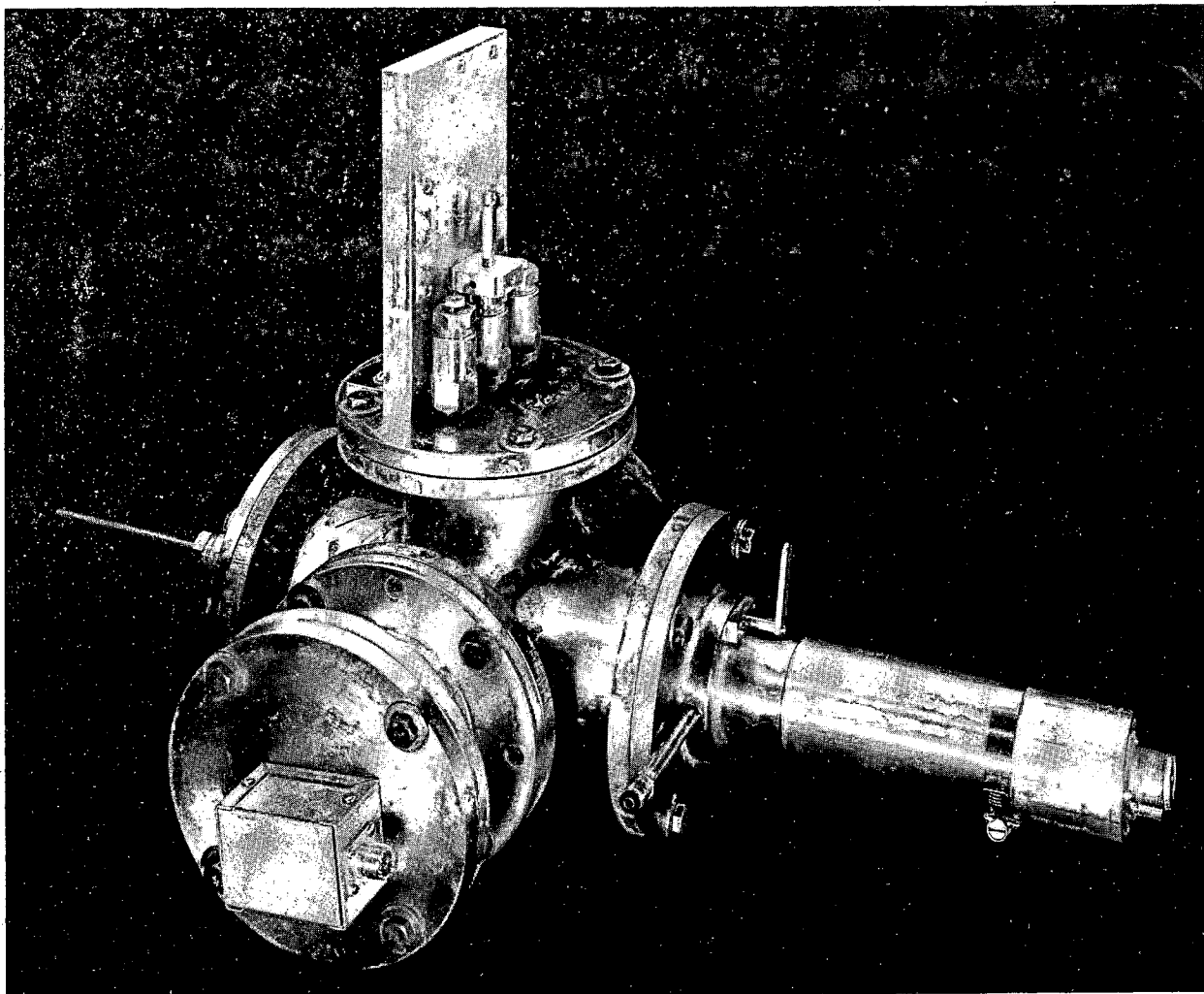
The experimental method of studying the properties of Faraday cups was similar to that used by Fulmer.¹²² The relative amount of beam that passed through a thin gold target (vaporized onto aluminum backing) was determined by counting the fission fragments emitted at 90 deg to the beam. Thus all the bombardments could be normalized to the same number of beam particles. In this way, it was possible to compare the indicated amount of charge collected for a given amount of beam with various positions and arrangements of the Faraday cup. The absolute calibration of the Faraday cup was done by counting elastically scattered beam particles at small angles to the beam. The measurements by Goldberg and Reynolds¹²³ indicate that at those angles the cross sections for elastic scattering are equal to those calculated by use of the Rutherford scattering equation.¹²¹

"T" Assembly

Many of the supplementary investigations were performed by using the "T" assembly shown in Fig. 43. At the entrance to the assembly, the beam was constrained by two 3/16-in-diam. collimators, 3 in. apart. The targets were attached to the center probe of the top plate. The gas-scintillation chamber and photomultiplier tube were attached to the side port at 90 deg to the beam direction. The lever shown just above the gas chamber (Fig. 43) was attached to a Cr^{252} sample inside the assembly. This made it possible to place the sample in front of the chamber window for energy calibrations, without letting the system up to atmospheric pressure. This assembly was used for the relative experiments. The absolute calibrations were done in the large vacuum tank described in Section II.

Faraday Cup

The Faraday cup used in the calibrations is shown schematically in Fig. 20 of Section II. In Fig. 43, only the electrical connector on a stationary, unshielded cup is shown (foreground). The copper Faraday



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Fig. 43. "T" assembly used in preliminary experiments.

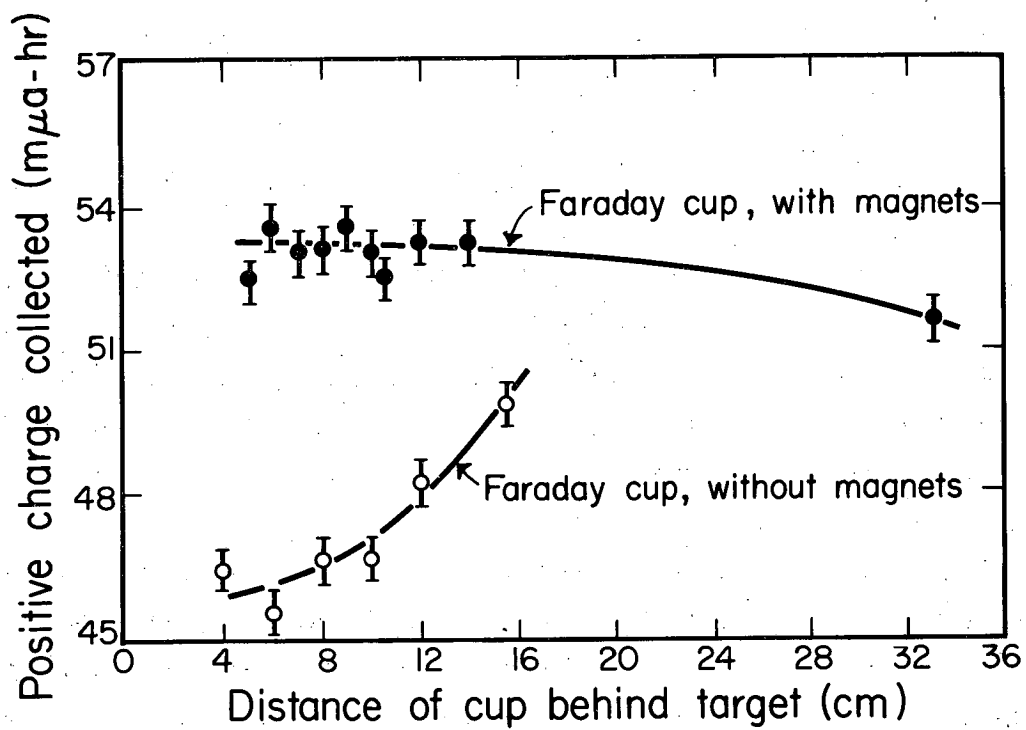
cup used in the calibration experiments was $1\frac{3}{4}$ in. long and $\frac{9}{16}$ in. in diameter. A set of bar magnets surrounded the cup and extended $1\frac{1}{8}$ in. in front of it. They maintained a magnetic field of 800 to 1200 gauss (perpendicular to the beam direction) in the region of the cup. The entire unit was attached to a probe which could be adjusted to place the cup at various distances behind the target.

Experimental Results

The results of the relative experiments are shown in Figs. 44 and 45. The upper curve of Fig. 44 gives the positive charge collected on the Faraday cup, with magnets attached, as a function of distance from the target. The lower curve shows the charge collected on the cup when the magnets have been removed. Figure 45 is a plot of the current observed on the magnets and shielding for a given positive current in the Faraday cup. Normally the magnets and shielding are grounded to prevent accumulation to electrostatic charge.

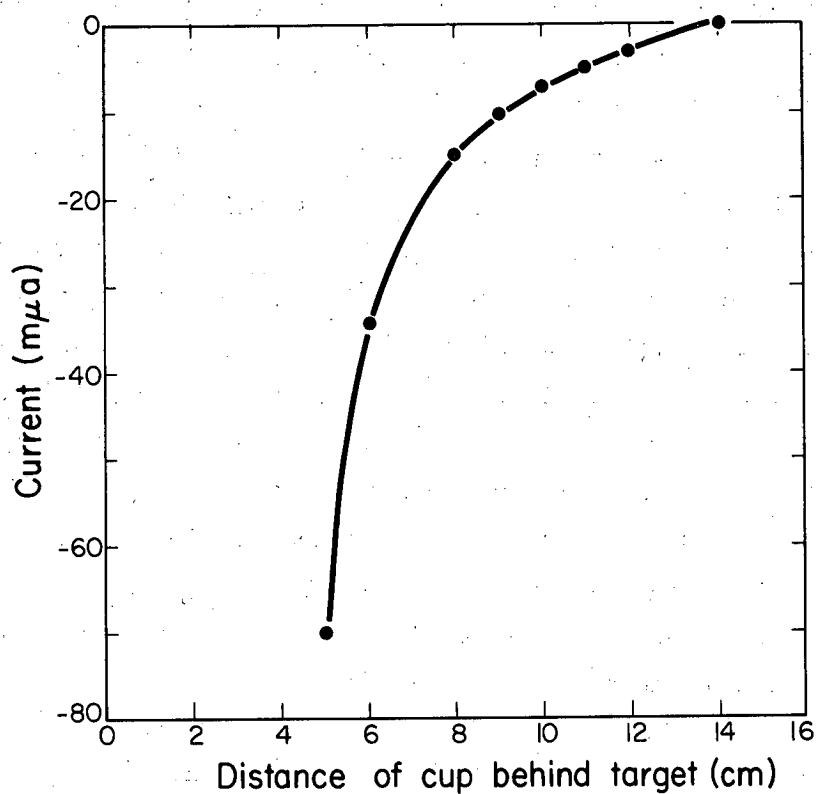
With magnets surrounding the cup, the amount of charge collected appears to decrease slightly as the cup is backed away from the target. This is apparently due to loss of particles by elastic scattering in the target. The error does not become serious until the cup is beyond about 15 cm behind the target. This scattering effect would, of course, be more serious for thicker targets. When the Faraday cup is near the target, the magnets and shielding, which extend $1\frac{1}{8}$ in. in front of the cup, receive large amounts of negative current. This is presumably due to electrons that have been knocked out of the target and been curved away from the Faraday cup. When the cup is backed away, the solid angle subtended decreases, thus lowering the number of electrons picked up for a given positive current in the cup. At 14 cm the current on the magnets and shielding becomes slightly positive, indicating that positive current due to scattered beam particles more than cancels the electron current.

When the magnets are removed from the cup, it appears that a considerable number of electrons are picked up, partially canceling the positive beam-particle currents. This effect decreases as the cup is backed away from the target.



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Fig. 44. Amount of positive charge collected to give 10^4 counts; various arrangements of Faraday cup.
Target: $206 \mu\text{g}/\text{cm}^2$ Au on $1.17 \text{ mg}/\text{cm}^2$ Al foil at 45° to beam, bombarded with full-energy C^{12} .



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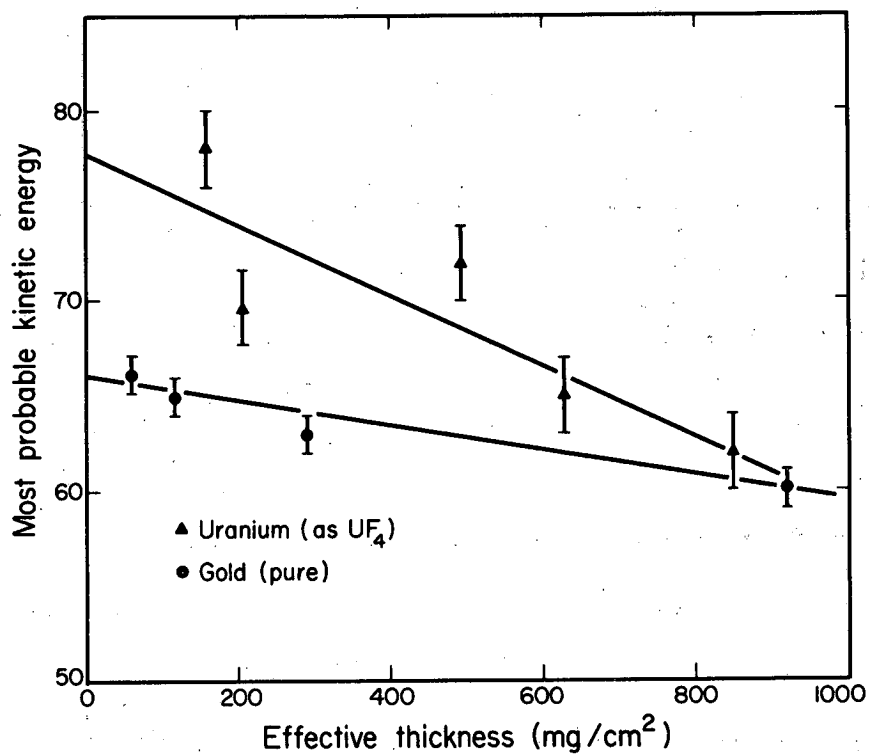
Fig. 45. Current picked up on shield and magnets for 300 mμa positive current in cup.
Target: 206 μg/cm² Au on 1.17 mg/cm² Al foil at 45 deg to beam, bombarded with full-energy Cl³⁵.

The absolute calibration of the magnetically shielded Faraday cup was done by counting elastically scattered C^{12} particles resulting from 72.4-Mev C^{12} bombardment of gold. Using the Ha-2 p-n junction, measurements were made at 10-deg intervals from 30 to 140 deg, where $\sigma_{\text{scatt}}/\sigma_{\text{Ruth}}$ drops to approx 0.01. A plot of $\sigma_{\text{scatt}}/\sigma_{\text{Ruth}}$ vs center-of-mass angle agreed quite well with the results of Goldberg and Reynolds for 73.6-Mev C^{12} on Au,¹²³ except that between 30 and 50 deg the ratio found in this experiment was 1.06 ± 0.04 . It is assumed that at these small angles the scattering cross section is equal to that calculated according to the Rutherford equation, and that the factor of 1.06 arises from an error in the Faraday cup reading. This discrepancy is in the direction indicating pickup of electrons or scattering of beam particles out of the solid angle subtended by the cup. Because of the small size of the cup and its magnetic shielding, the latter explanation is much more likely. Faraday cup readings obtained in absolute fission-fragment counting experiments have been corrected for this error. In arriving at the factor of 1.06, account has been taken of the fact that the average equilibrium charge of C^{12} particles of this energy is 5.97.¹²⁴ Also, scattering cross sections obtained at forward angles have been corrected for the finite angular width of the detector.¹²⁵

2. Target Thickness Effects

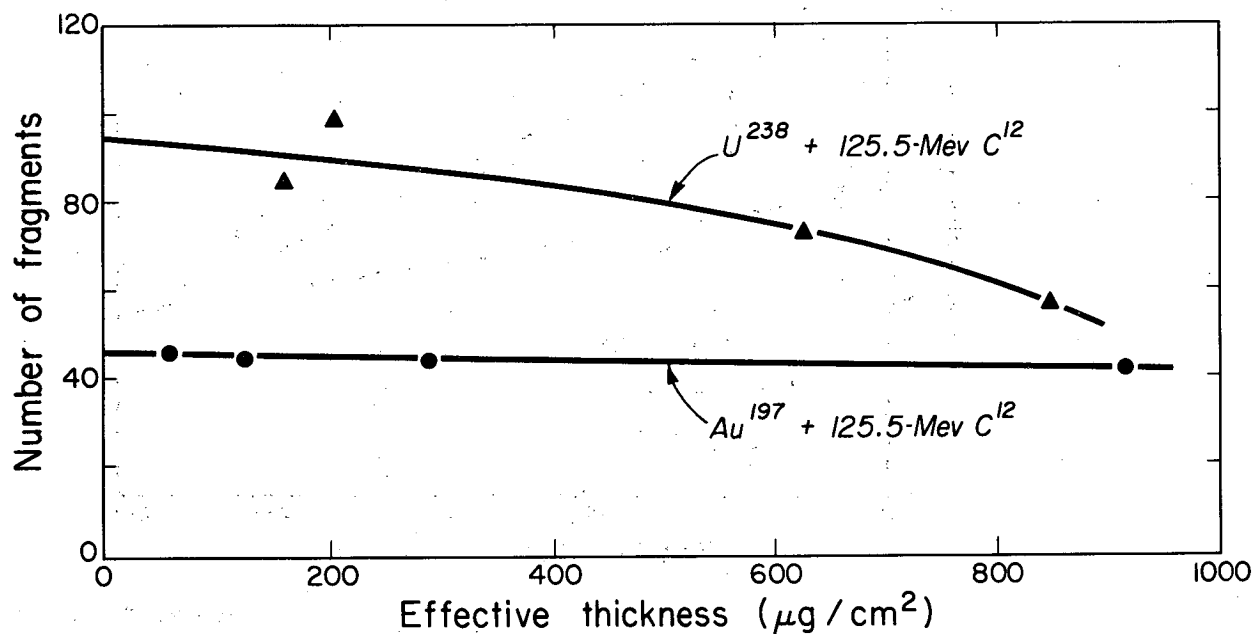
In order to be able to correct for kinetic energy loss and absorption of fragments in the target material, targets of various thicknesses were bombarded and the fission-fragment spectra were observed at 90 deg to the beam in the gas scintillation chamber. These experiments were performed in the T assembly described in the preceding section of this Appendix.

The results obtained are shown in Figs. 46 and 47. In Fig. 46, the most probable fragment kinetic energy is plotted as a function of effective thickness of the fissionable material. The term "effective thickness" means the target thickness with respect to the counter. In this case, the effective thickness was $\sqrt{2}$ times the actual thickness because the targets were at 45 deg to the beam and the counter was at 90 deg. The kinetic energies were determined from the positions of the peaks of the fragment kinetic energy spectra in the manner described in Section II.



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Fig. 46. Most probable fragment kinetic energy as a function of effective thickness of fissionable material for gold (pure) and uranium (as UF₄) bombarded with 123-Mev C¹² ions.



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Fig. 47. Number of fragments observed (at 90 deg to beam) per $\mu\text{a-hr}$ per $100 \mu\text{g}/\text{cm}^2$ effective thickness of fissionable material as a function of effective thickness.
 Targets: UF_4 on 0.03-mil N;
 Au on 0.1-mil Al.

For the pure gold targets, the kinetic energy is not greatly reduced over the range of thicknesses studied. It appears, however, that the presence of fluoride ions on the uranium target causes the kinetic energy loss to be roughly three times as great as for the same thickness of gold on a target. This is in qualitative agreement with the results of Alexander and Gallagher.¹²⁶ The curves for kinetic energy vs thickness have been used to correct the kinetic energies obtained at various angles and bombarding energies for loss in passing through the target material. Admittedly, this one set of results should not be used for all corrections. At forward angles, the fission fragments have higher energies and would be expected to lose more energy in the target than at 90 deg or at backward angles.^{71,127,128} However, since the corrections were small for all targets used to obtain kinetic energies, I did not consider it worth while to investigate the target-thickness effect at all angles.

Figure 47 shows the number of fragments observed at 90 deg per unit effective thickness (to the beam) per unit beam as a function of effective target thickness (to the counter). If there were no stopping of fragments in the target material or preferential scattering in or out of the solid angle subtended by the counter, this would be a constant. Over the range of thicknesses studied, the number of fragments from the gold targets is constant within experimental error. However, the number of fragments from the UF_4 targets drops off seriously with increasing thickness. This is apparently another manifestation of the greater stopping power of the light elements, as shown above in the kinetic energy experiments. No corrections for number of fragments as a function of target thickness have been applied to the angular distributions obtained with the gold targets as, in all cases, their effective thicknesses have been less than $300 \mu\text{g}/\text{cm}^2$. If angular distributions were obtained by using UF_4 targets, it would be necessary to use much thinner targets in order to minimize these effects.

APPENDIX B. Fission Model Calculations

1. Transformations

a. Introduction

In order to theoretically interpret the data from the fission counting experiments it is necessary to transform the angular distributions and kinetic energies from the laboratory system to the center-of-mass system. Because of the nature of the fission process, with its distributions of energies and masses, it is not possible to make an exact transformation when the data are obtained with a single counter. It has thus been necessary to make simplifying assumptions about the distribution of masses and the variation of total kinetic energy release with mass ratio. In order to test the validity of the assumptions made in Section II, calculations based on a plausible model have been made. The results of these calculations are discussed in the following paragraphs.

b. General Transformations

The calculations are based on a general description of the fission reaction similar to that previously used by several authors.^{28,88,105,129,130} Fission is pictured as occurring from a compound system moving with a velocity v_{comp} in the direction of the beam. It is assumed that the velocity vector of the compound system has no component perpendicular to the beam direction. The velocity-vector diagram of the fission process (shown for one of the fragments only) is given in Fig. 48.

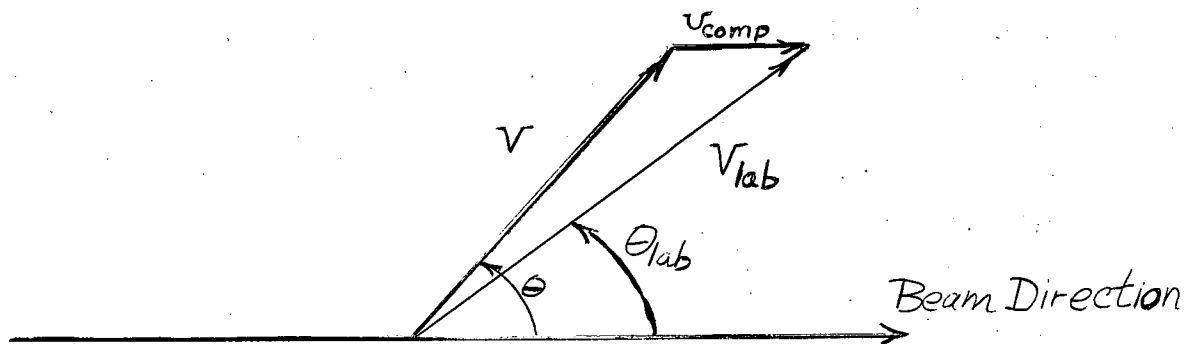


Fig. 48. Velocity-vector diagram of fission process.

The kinetic energy in the laboratory system, E_{lab} , of a fragment of energy E and velocity V in the center-of-mass system, emitted at a center-of-mass angle θ , is given by

$$E_{\text{lab}} = E(1 + \eta^2 + 2\eta \cos\theta), \quad (\text{B1})$$

where $\eta = v_{\text{comp}}/V$.

The laboratory-system and center-of-mass system angles are related by the expression

$$\tan\theta_{\text{lab}} = \frac{\sin\theta}{\eta + \cos\theta}. \quad (\text{B2})$$

In transforming an angular distribution from one system to the other, it is necessary not only to change the angles as indicated in Eq. (B2), but also to apply a correction factor for the change in solid angle. If we have

$$(d\sigma/d\omega)_{\text{cm}} = (d\sigma/d\omega)_{\text{lab}} G(\eta, \theta_{\text{lab}}), \quad (\text{B3})$$

where $(d\sigma/d\omega)_{\text{cm}}$ and $(d\sigma/d\omega)_{\text{lab}}$ represent the differential cross section per unit solid angle in the center-of-mass and laboratory systems, then the solid-angle correction factor, $G(\eta, \theta_{\text{lab}})$, is given by

$$G(\eta, \theta_{\text{lab}}) = \frac{[1 - \eta^2 \sin^2\theta_{\text{lab}}]^{1/2}}{[\eta \cos\theta_{\text{lab}} + (1 - \eta^2 \sin^2\theta_{\text{lab}})^{1/2}]^2}. \quad (\text{B4})$$

The derivation of this result plus much more useful information about the transformations is given in a report by Marion, Arnette, and Owens.¹³¹

c. Mass-Yield Distributions

The mass-yield data of Fairhall for 22-Mev deuteron-induced fission of Bi^{209} are very closely approximated by a Gaussian function having a full-width at half maximum of 16 mass units and centered at mass 103.¹³² A similar type of distribution, at least in the mass region of high yields, has been found for the fission of gold with 115-Mev N^{14} .³⁷ In

these calculations a Gaussian mass-yield distribution of full width at half maximum of 16 mass units is assumed for the fission of Au^{197} with 120-Mev C^{12} ions.

d. Total Kinetic Energy Release

The variation of total kinetic energy release with mass ratio of the fragments is probably the least well known of the functions needed for these calculations. One could, as a first approximation, assume that over the small region of masses produced in high yield the total kinetic energy release is constant. However, it is probably more meaningful to assume that the fragment kinetic energies arise from mutual Coulomb repulsion of the fragments following scission. According to this model, the fragments are pictured as cotangential charged spheres and the kinetic energy release is equal to the electrostatic potential energy of the spheres in contact. If one assumes that the charge-to-mass ratio is the same for each of the fragments, the average total kinetic energy release for various fissioning nuclei is proportional to $Z^2/A^{1/3} r_0$. Terrell has rather successfully correlated fission kinetic energy release with $Z^2/A^{1/3}$; however, he notes that the value of r_0 needed to fit the experimental data is at least 25% greater than the radius parameter obtained from other types of experiments.²² Some of this difference is probably due to distortion of the fragments and a tendency for protons of the two fragments to be separated more than the neutrons, but he states that some of the increase is likely due to expansion of the highly excited fragments.

For a given fissioning nucleus, the variation of average total kinetic energy release with mass ratio is in fair agreement with this model, although for fission in the region of uranium and above there are definite indications that there is a decrease in kinetic energy release as a mass ratio of unity is approached.^{43,106,107} No data have been reported on the variation of kinetic energy release with mass ratio in the region of astatine compound nuclei. For this reason, and for simplicity, it is assumed that the $Z_1 Z_2 / (A_1^{1/3} + A_2^{1/3})$ energy variation is obeyed.

In addition to the kinetic energy variations discussed above, it has been observed that even for a given ratio of masses there is a distribution of kinetic energies rather than a unique energy release for each mass ratio.^{43,106,107} It is assumed in these calculations that this distribution is Gaussian, with a full width at half maximum of about 15 Mev. This would seem to be a reasonable assumption on the basis of the experimental results mentioned above.

e. Calculations

The calculations have been made for the system $\text{Au}^{197} + 120\text{-Mev C}^{12}$. It is assumed that

- (i) The sum of the mass numbers of the fission fragments is 200.
- (ii) The most probable total kinetic energy release for symmetric division is 140 Mev.
- (iii) The momentum vector of the compound nucleus equals that of the bombarding particle.

The equation for the assumed Gaussian mass-yield distribution is

$$P(A) = C \exp [-k_1 (A_0 - A)^2], \quad (\text{B5})$$

where $P(A)$ = probability of formation of a fragment of mass number A ,

A_0 = the mass number of the most probable fragment, and

C is a normalizing constant. The parameter k_1 is given by

$$k_1 = \frac{4 \ln 2}{a^2}, \quad (\text{B6})$$

where a = the full width of the mass-yield distribution at half maximum.

In keeping with the assumptions made above, the most probable total kinetic energy release, $\bar{E}_T$, as a function of the fragment masses and charges is given by

$$E_T = K \frac{Z_1 Z_2}{A_1^{1/3} + A_2^{1/3}}. \quad (\text{B7})$$

(Note: The constant K has the units of e^2/r , or energy. Its absolute value could be calculated from first principles if the model of Coulomb repulsion of charged spheres were strictly invoked. However, owing to the deviations from this model that were discussed above, K has been left as a parameter, and adjusted to give an $\bar{E}_T$ of 140 Mev for symmetric fission from the Au + C¹² system.)

If it is assumed that the charge-to-mass ratio is the same for each of the fragments of any given event, $\bar{E}_T$ is given in terms of the mass number of one of the fragments by

$$\bar{E}_T = K \frac{A_1(2A_0 - A_1)}{A_1^{1/3} + (2A_0 - A_1)^{1/3}} \quad (B8)$$

In the system of the fissioning nucleus, the magnitudes of the momenta of the two fragments are equal. Imposing this condition upon Eq. (B8), one obtains the following expression for the most probable kinetic-energy of Fragment 1:

$$\bar{E}_T = \frac{K'}{2A_0} \frac{A_1(2A_0 - A_1)^2}{A_1^{1/3} + (2A_0 - A_1)^{1/3}} \quad (B9)$$

For each fragment mass there is a Gaussian kinetic energy probability distribution centered about $\bar{E}_1$. Thus for a fragment of mass number A_1 , the probability of its having a kinetic energy between E_1 and $E_1 + dE_1$ is equal to $P_{A_1}(E_1)dE_1$, where

$$P_{A_1}(E_1) = C' \exp [-k_2(\bar{E}_1 - E_1)^2], \quad (B10)$$

$\bar{E}_1$ is given by Eq. (B9), C' is a normalizing constant, and k_2 is a function of the width of the energy distribution. It is assumed that the sum of the full widths at half maximum of the energy distributions of complementary fragments A_1 and A_2 is 20 Mev, and that the width of each individual distribution is proportional to its most probable energy. From these assumptions we have

$$k_2 = \frac{4 \ln 2}{(10A_0/A_1)^2} \quad (B11)$$

The mass-yield and kinetic energy distributions have been combined to give a combined probability surface. The probability of occurrence of a fragment of mass number between A and $A + dA$, and of kinetic energy between E and $E + dE$, is proportional to $P(E,A)dEdA$, where

$$P(E,A) = CC' \sqrt{k_2} \exp [-k_1(A_0 - A)^2] \exp [-k_2(\bar{E} - E)^2]. \quad (B12)$$

This expression is the result of combining Eqs. (B5) and (B10). It is necessary to include the factor $\sqrt{k_2}$ in order to make the integrated probabilities of complementary fragments equal. The constants CC' have been adjusted to yield a probability of unity for symmetric division ($A = 100$) with an energy per fragment of 70 Mev. The probability surface is represented by a contour diagram in Fig. 49. In order to avoid confusion, only a few of the calculated contour lines are shown.

From the assumption of full momentum transfer by the bombarding particle to the compound nucleus, the value of η^2 is a function only of the ratio A/E of the fragment, and is given by

$$\eta^2 = \frac{A_{C12} E_{C12}}{A_{\text{comp}}^2} \left(\frac{A}{E} \right). \quad (B13)$$

Lines of constant η^2 have been included in Fig. 49.

It is easier to obtain the probability distribution of η if one first transforms the probability surface of Fig. 49 to cylindrical coordinates, i.e.,

$$E = r \sin \theta; A = r \cos \theta; \text{ and } P(E,A) = z. \quad (B14)$$

(Note: The angle θ used here is not to be confused with the θ 's that represent angle with respect to the beam direction.)

In this representation, the lines of constant η^2 are simply lines of constant θ , and the relative probability of occurrence of a value between η^2 and $\eta^2 + d(\eta^2)$ is $P(\eta^2)d(\eta^2)$, where

$$P(\eta^2) \propto \sin^2 \theta \int P(E,A) r dr, \quad (B15)$$

where θ is constant and given by the relation

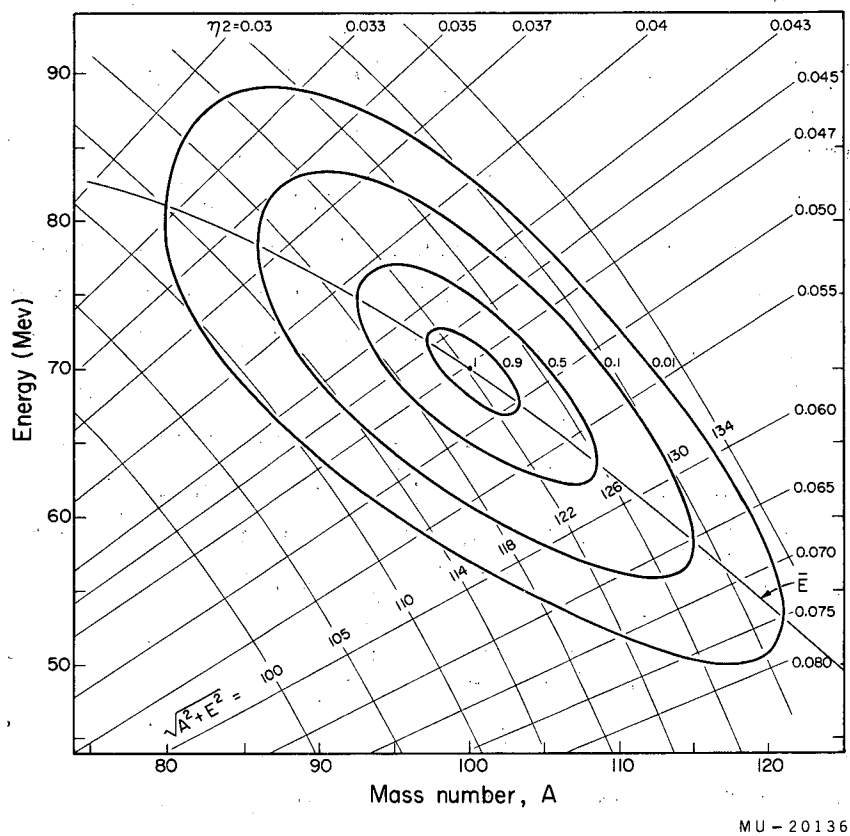


Fig. 49. Contour diagram of the probability surface, $P(E, A)$.

$$\eta^2 = \frac{A_{C12} E_{C12}}{A_{comp}^2} \cot \theta. \quad (B16)$$

The factor $\sin^2 \theta$ arises from

$$\frac{d(\eta^2)}{d\theta} = \frac{-1}{\sin^2 \theta} \frac{A_{C12} E_{C12}}{A_{comp}^2}. \quad (B17)$$

When substitutions are made in Eq. (B15) from the transformations of (B14), the expression for $P(\eta^2)$ becomes

$$P(\eta^2) \propto \frac{E^2}{E^2 + A^2} \int_0^{\left(\sqrt{A^2 + E^2}\right)_{\max}} P(E, A) \sqrt{A^2 + E^2} d(\sqrt{E^2 + A^2}), \quad (B18)$$

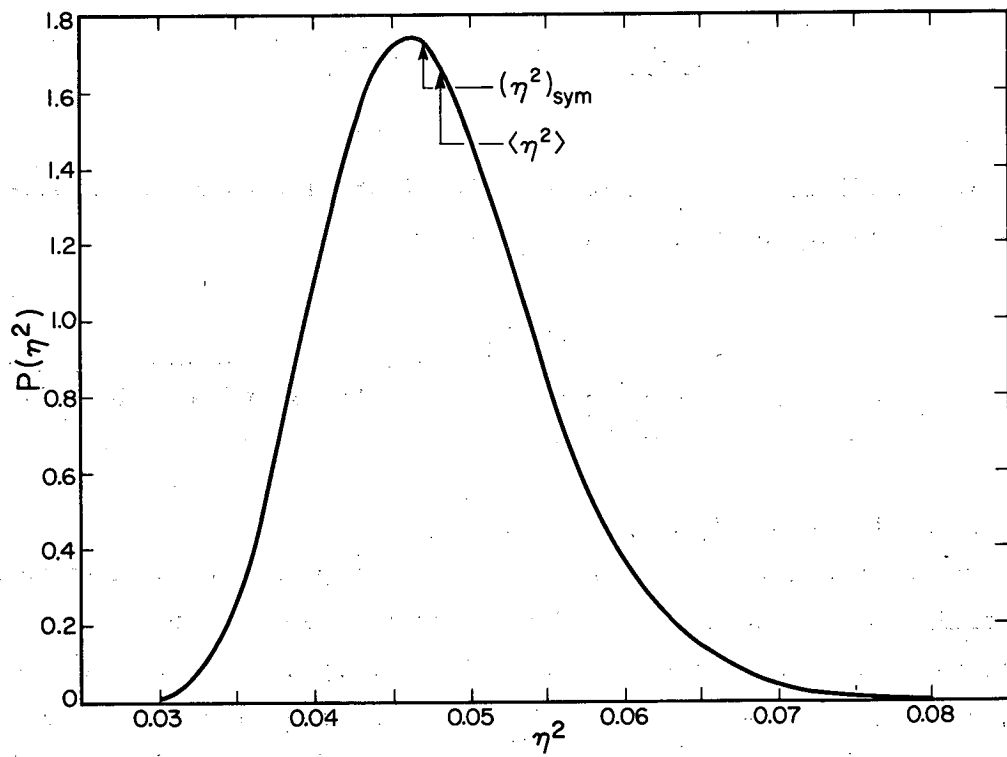
where the ratio A/E is given by Eq. (B13).

The probability distribution for η^2 was obtained by graphically performing the integrations of Eq. (B18) for many values of η^2 . In practice, the integration was carried out only in the regions in which $P(E, A) \geq 0.005$. The resulting distribution of η^2 is shown in Fig. 50. The average value $\langle \eta^2 \rangle$ was obtained graphically according to the equation

$$\langle \eta^2 \rangle = \frac{\int \eta^2 P(\eta^2) d(\eta^2)}{\int P(\eta^2) d(\eta^2)}. \quad (B19)$$

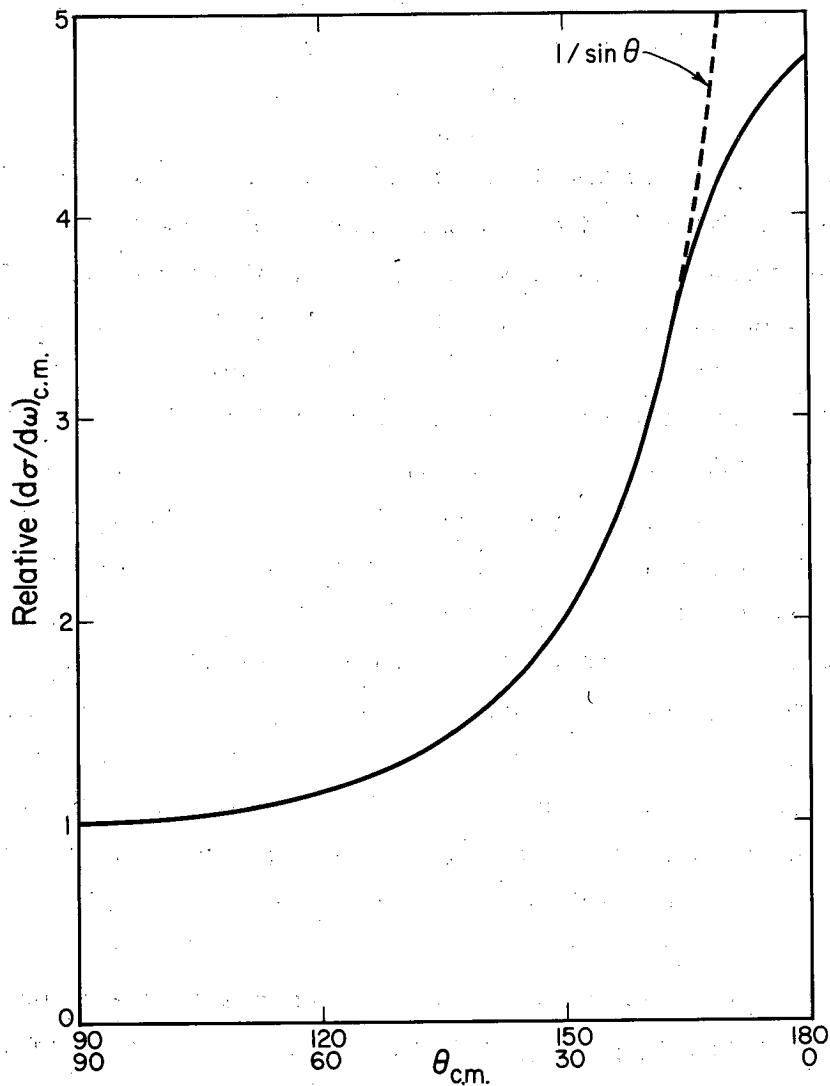
It is to be observed that $\langle \eta^2 \rangle$ is slightly greater than the value of η^2 corresponding to the most probable mode of symmetric fission $(\eta^2)_{\text{sym}}$.

The distribution of η^2 was then used to investigate the transformation of angular distributions between the center-of-mass and laboratory systems. The center-of-mass angular distribution shown in Fig. 51 was assumed for all fragments, regardless of η value. The assumed angular distribution is approximately the same as that observed for 123-Mev C^{12} fission of Au^{197} (see Section IV). This angular distribution was transformed to the



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Fig. 50. Probability distribution of η^2 .



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Fig. 51. Assumed center-of-mass angular distribution (for all η values).

laboratory system by using η^2 values of 0.03, 0.03, ..., 0.08. The resulting angular distributions in the laboratory system for each value of η^2 were multiplied by

$$\int_{\eta^2-0.005}^{\eta^2+0.005} P(\eta^2) d(\eta^2),$$

and summed to obtain the total angular distribution shown in Fig. 52.

This angular distribution was then converted back to the center-of-mass system by using the single value of η^2 equal to $\langle \eta^2 \rangle$. Comparison of the resulting curve, shown in Fig. 53, with the input center-of-mass angular distribution of Fig. 51 indicates that, to a very good approximation, transformation of the total angular distribution from the laboratory system to the center-of-mass system by using the average value of η^2 faithfully reproduces the center-of-mass distribution for the wide range of η values. Of course, this will not be true if the center-of-mass distributions are different for different values of η . However, Knox et al. have analyzed their results in the $\text{Au}^{197} + \text{O}^{16}$ system by assuming a constant fragment mass and dividing the energy spectra into various energy groups, and obtaining a center-of-mass angular distribution for each energy group.⁹⁹ Within the limits of error, all the angular distributions are the same, adding support to the assumption of the same angular distribution in the center-of-mass system for groups of different η values.

The kinetic energy spectrum of fragments in the center-of-mass system is obtained by integrating $P(E,A)dA$ along lines of constant E . The resulting spectrum is shown in Fig. 54. Note that the peak of this distribution corresponds to the most probable kinetic energy per fragment assumed for symmetric division. It has been assumed in Section II that in the laboratory system the most probable kinetic energy at a given angle corresponds to the energy per fragment upon symmetric division. Because of the great number of graphical integrations necessary, no attempt has been made to transform the kinetic energy spectrum to various angles of the laboratory system. However, rough calculations indicate that the assumption remains valid when the transformations are performed.

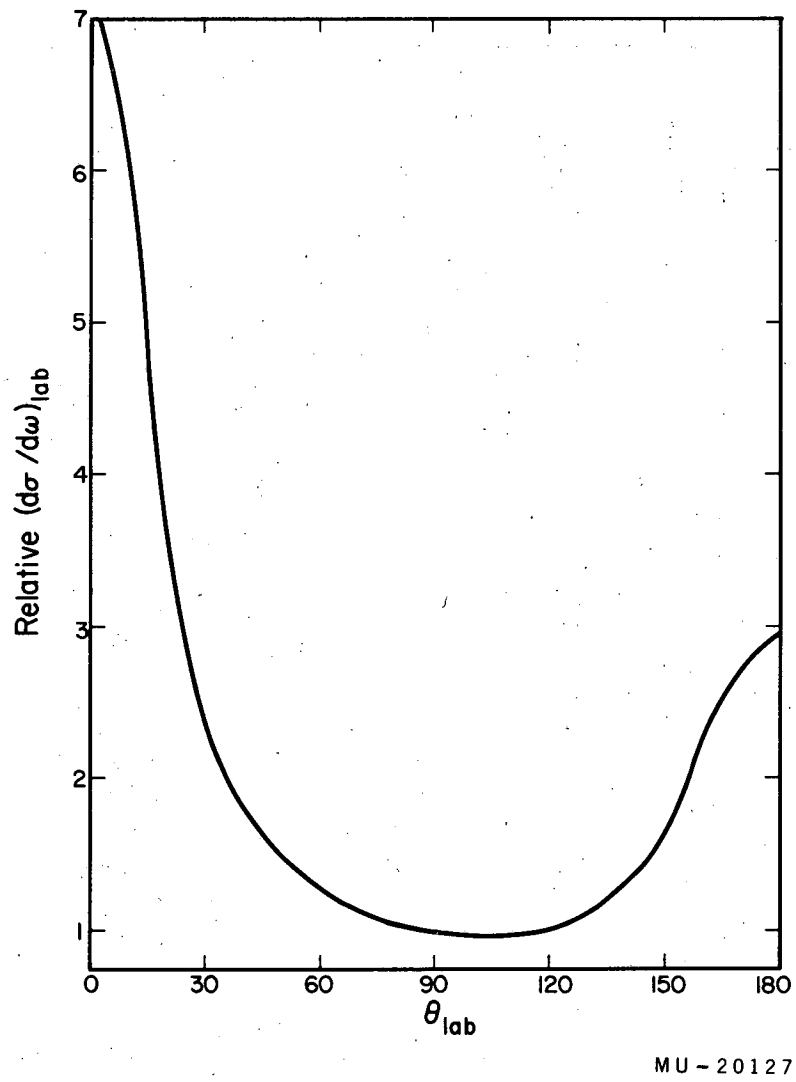
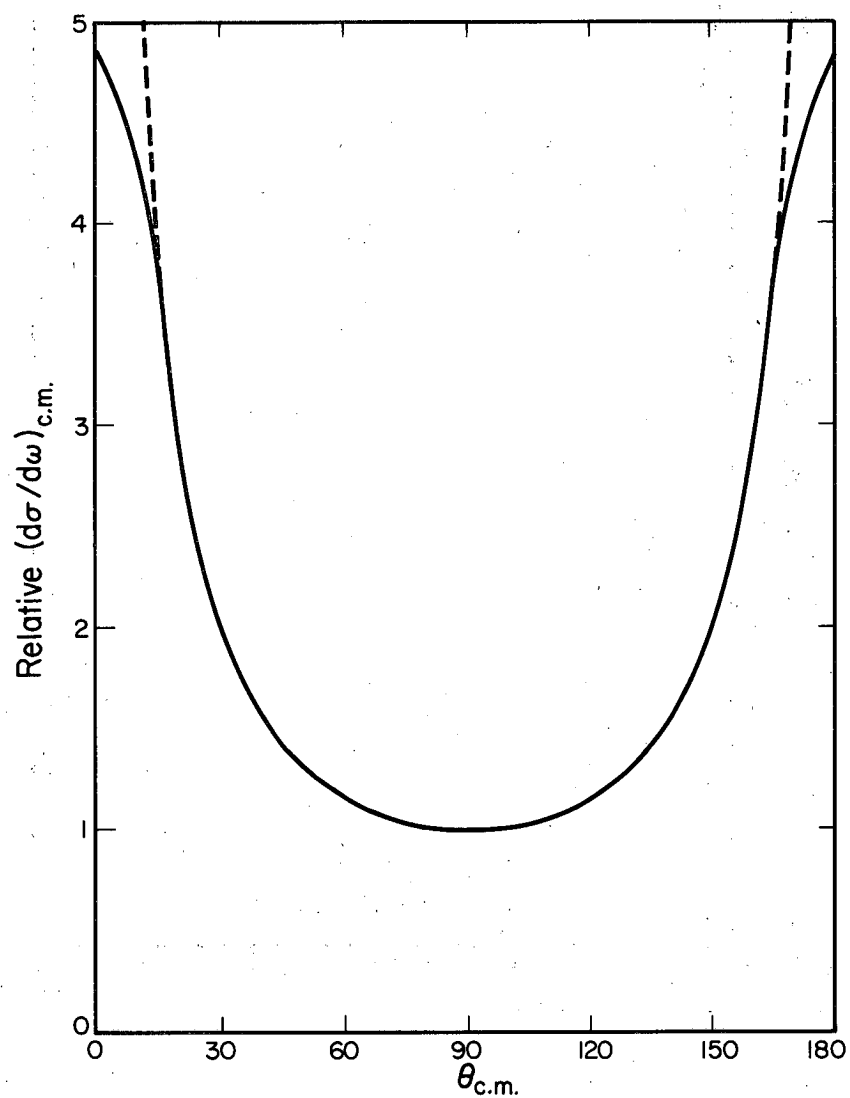
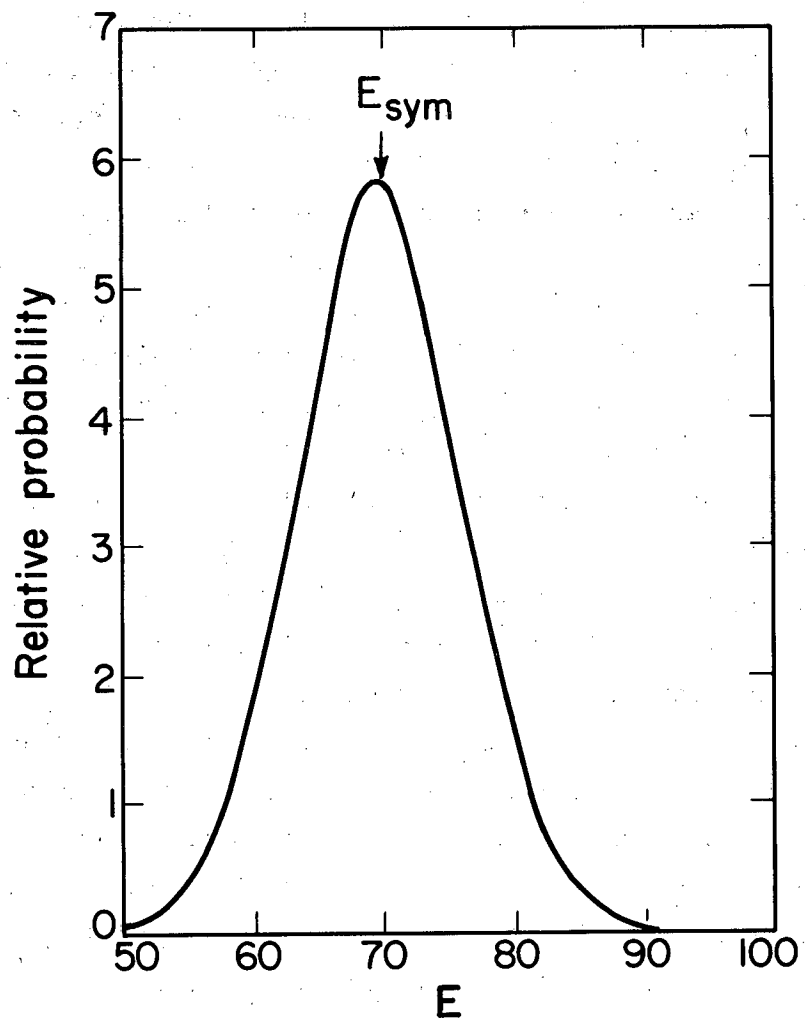


Fig. 52. Total angular distribution, laboratory system.



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Fig. 53. Center-of-mass angular distribution obtained by transformation of total angular distribution (Fig. 52), using $\langle \eta^2 \rangle$.



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Fig. 54. Fragment kinetic energy spectrum, center-of-mass system.

f. Other Factors

Because of the large number of variables present in beam-induced fission it has been feasible to treat only those considered most important. However, it should be pointed out that there are several others which would have to be considered if a rigorous analysis were attempted.

The effects of neutrons have been neglected except for the assumption that the sum of the fission-fragment masses is nine units less than the mass of the compound nucleus. Also, the emission of heavy particles from the compound nucleus prior to fission has been ignored. This process could have the effect of giving the compound nucleus a velocity component perpendicular to the beam direction, whereas it was assumed that we have $\vec{p}_{\text{comp}} = \vec{p}_{\text{C}^{12}}$. Even the assumption of formation of a compound nucleus may not be justified in all cases. For example, Alexander¹⁰⁰ has found evidence for fission from non-compound-nucleus reactions in the system $\text{U}^{238} + \text{C}^{12}$. However, as discussed in Section IV, the experiments of this work in the $\text{Au}^{197} + \text{C}^{12}$ system indicate full momentum transfer by the bombarding particle to the fissioning system, implying formation of a compound nucleus.

No mention has been made of the possibility of ternary fission, i.e., division of the nucleus into three massive fragments. This is not expected to be an important process in this region of compound nuclei. (Note added in preparation: Recent information from H. M. Blann indicates that the full width of the mass-yield distribution at half maximum is approx. 27 mass units for fission of gold with 120-Mev C^{12} particles.¹¹⁶ This is considerably larger than the value assumed in the calculations in this section, and would broaden the probability distributions. However, it would not change any of the conclusions drawn from results of the calculations.)

2. Level-Density Calculations

According to the calculations of Ericson and Strutinski,¹¹³ the density of levels of spin j in a compound nucleus having excitation energy E and nuclear temperature T is given by

$$\rho_j = \rho_0 (2j+1) \exp[-h^2 j(j+1)/2\mathfrak{I}T] \quad (\text{B20})$$

where ρ_0 = density of levels of zero spin. The nuclear moment of inertia, $\mathfrak{I}$, is assumed to be equal to the rigid-body moment of inertia ($= \frac{2}{5} mAR^2$).

This equation may be derived from two different approaches. On one hand, the functional form of Eq. (B20) is obtained on very general grounds if $\vec{j}$ is the sum of a large number of component vectors oriented at random in space. This is simply a random-walk problem. The actual expression for ρ_j given in Eq. (B20) with constants was obtained by the assumption that collective rotational energy of a nucleus is not available for intrinsic excitations. This assumption gives rise to the Boltzman factor involving the rotational energy, $\exp[-h^2 j(j+1)/2\mathfrak{I}T]$.

In order to obtain a quantitative notion of the importance of the effects of level densities upon spallation reactions, calculations were made according to Eq. (B20). The results of these calculations are given in Table B-I. In making the calculations, it was assumed that $T = (10E/A)^{1/2}$.

Table B-I. Densities of levels of various spins

E(Mev)	Most probable spin, $\mathfrak{I}_{m.p.}$	ρ_5/ρ_0	ρ_{10}/ρ_0	ρ_{20}/ρ_0	ρ_{40}/ρ_0
5	8.3	9.1	10.4	2.87	0.0024
10	10	9.6	12.8	6.2	0.051
15	11.1	9.9	14.0	8.8	0.194
20	12	10	14.8	10.8	0.445
40	14.4	10.3	16.4	16.0	2.02
60	16	10.4	17.2	19.2	4.05

Note that (1) the most probable spin value is an increasing function of energy and (2), at the lower excitation energies, ρ_j/ρ_0 drops off rapidly with increasing j above $j_{m.p.}$. There is some question as to the proper value to use for S . The large body of data obtained from the study of ground-state rotational bands of spheroidally deformed nuclei indicates that S is much smaller than the rigid-body value. If the lower value of S were used in Eq. (B20) the level-density distributions would be shifted to lower values of j and a more drastic difference between distributions at low excitation energy and those at high energy would be obtained.

APPENDIX C. Decay Properties of the Astatine Isotopes

1. Introduction

In order to accurately determine the amounts of the various astatine isotopes produced in the $Pt + N^{14}$ nuclear reactions, it was necessary to know their half lives and alpha/EC branching ratios. In many cases, the branching ratios had not been determined. Thus it was necessary in the course of this work to determine some of these values. The alpha-branching ratios were determined by observation of the number of alpha particles emitted by the astatine isotopes and by their polonium daughters. In most cases, these data were obtained as by-products of the cross-section experiments and not from experiments designed specifically for the study of decay properties. During the progress of this work, similar investigations were made by Latimer and Thomas.³⁵ The final values used for the branching ratios were obtained by comparison with their results and by selection of either the average value or the one determined with the highest accuracy.

The values used for the half lives, alpha-particle energies, and percentage decay by alpha emission are listed in Table C-I. In Table C-II are listed the decay properties of the polonium isotopes, upon which most of the astatine alpha-branching values are based. More detailed accounts of some of the investigations are given in the following paragraphs.

Table C-I. Decay properties of the astatine isotopes

Isotope	Half-life	Ref.	E_{α} (Mev)	Ref.	Alpha branching (%)	Ref.
At ²⁰²	3.0±0.2 min	32	6.231 (36%) 6.133 (64%)	32	12.0±0.4	a
At ²⁰³	7.4±0.3 min	32	6.086	32	13.8±0.3	35
At ²⁰⁴	9.3±0.2 min	32	5.950	32	4.4±0.2	a
At ²⁰⁵	26.2±0.5 min	32	5.899	32	18.4±0.6	a
At ²⁰⁶	29.5±0.6 min	a	5.699	32	0.88±0.08	a
At ²⁰⁷	107.8±2.7 min	a	5.750	32	10	133
At ²⁰⁸	1.6±0.2 hr	134	5.65	141	0.555±0.055	141
At ²⁰⁹	5.5 hr	133	5.642	134	~5	133
At ²¹¹	7.20 hr	143	5.89	133	40.9	142

^aDetermined in this work with collaboration of Latimer and Thomas.³⁵

Table C-II. Decay properties of the polonium isotopes

Isotope	Half-life	Ref. ^a	E (Mev)	Ref. ^a	Alpha branching (%)	Ref. ^a
Po ²⁰²	51±3 min	134	5.575		2	134
Po ²⁰³	45 min	145	5.48	145	---	
Po ²⁰⁴	212±2 min	b	5.370		0.63	120
Po ²⁰⁵	1.8 hr		5.21±0.01		0.074±0.016	146
Po ²⁰⁶	8.8 da	136	5.218	134	5±1	137
Po ²⁰⁷	5.7 hr		5.10±0.02		0.014	147
Po ²⁰⁸	2.93±0.03 yr		5.109		100	
Po ²⁰⁹	103 yr		4.86		99+	
Po ²¹¹	0.52 sec		7.442		100 (β-stable)	

^aWhere specific references have not been given, references to the original literature may be found in Ref. 144.

^bDetermined in this work with collaboration of Latimer and Thomas.³⁵

2. At²⁰⁷

Astatine-207 was first reported by Barton, Ghiorso, and Perlman, who found the half life to be approx 2 hr.¹³³ From the counting rate of the electron-capture daughter, Po²⁰⁷, and the number of alpha particles emitted by At²⁰⁷, they estimated the alpha branching to be about 10%. As the authors point out, there may be considerable uncertainty in this value, as the counting efficiency of the Po²⁰⁷ radiations was not precisely known. Later reports list the half life of At²⁰⁷ as 107±5 min¹³⁴ and 108±5 min.¹³⁵ The energy of the alpha particles was found by Hoff, Asaro, and Perlman to be 5.750 Mev.³²

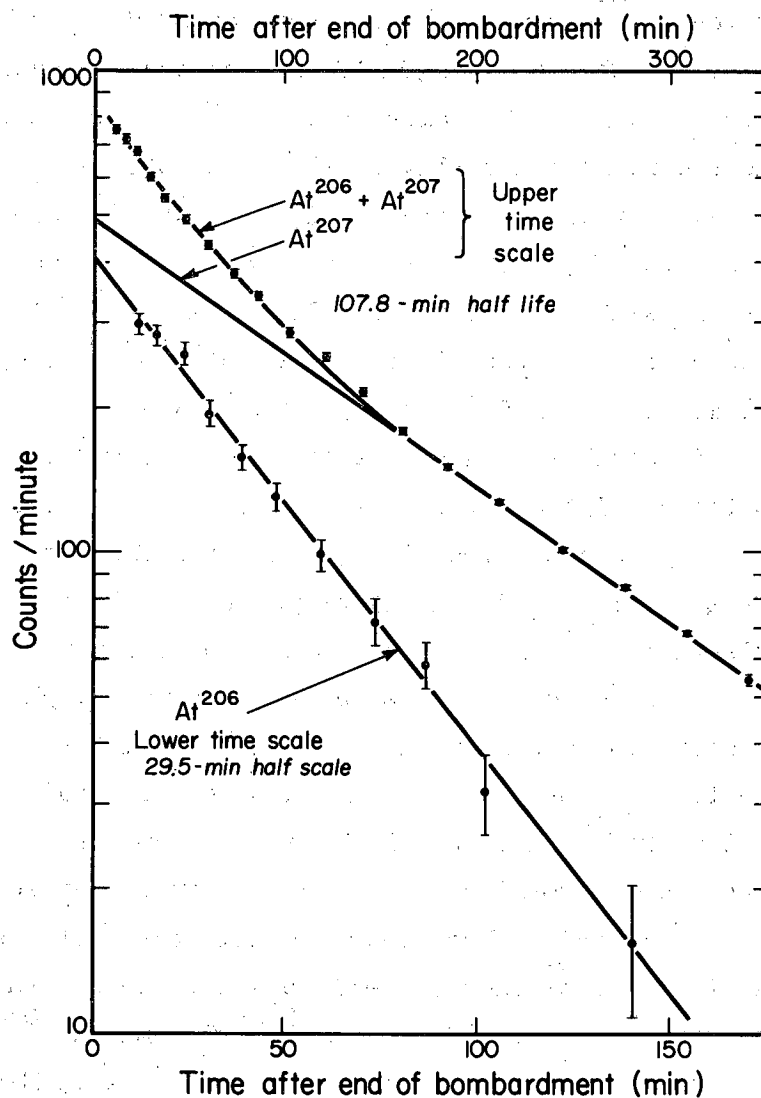
The half life for At²⁰⁷ determined in this work is 107±2.7 min. This value was obtained by a least-mean-square fit of the decay of the 5.75-Mev alpha group of several of the better experiments (note Fig. 55).

3. At²⁰⁶

Barton, Ghiorso, and Perlman have reported a 2.6-hr. half life for this isotope, which decays by electron capture.¹³³ No direct radiations from At²⁰⁶ were observed, but the half life was determined by separating and counting the alpha-particle radiations of its daughter, Po²⁰⁶. This work was substantiated by Stoner, who found the half life to be 2.9±0.4 hr.¹³⁴ The alpha-decay branching is thought to be extremely small, as no alpha group with that half life has been observed.

Hoff, Asaro, and Perlman recently reported an alpha group of 5.699 Mev energy from At²⁰⁶ which decays with a half life of 21.7±2.6 min.³² This isotope was produced by the reaction Au¹⁹⁷(C¹²,3n)At²⁰⁶. They report no evidence for production of the 2.6-hr activity.

In this study, the resolution of the alpha-particle pulse-height analyzer was not good enough to separate the 5.699-Mev group emitted by At²⁰⁶ from the 5.75-Mev particles of At²⁰⁷. Thus the combined activities were observed as a function of time and resolved by analysis of the decay curve. An example of the decay curves obtained is shown in Fig. 55. A least-squares analysis of several of the curves for which the statistics were good yielded a value of 29.5±0.6 min for the At²⁰⁶ half life. This



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Fig. 55. Resolution of the decay curve of At²⁰⁶ and At²⁰⁷ alpha activities.

value has been used in calculating the reaction cross sections.

The alpha-branching ratio was determined by counting the alpha particles of Po^{206} several hours after isolation of the astatine. In calculating the At^{206} branching ratio, values of 8.8 days¹³⁶ and 5%¹³⁷ have been used for the Po^{206} half life and percentage decay by alpha emission, respectively. The value obtained is $0.88 \pm 0.08\%$.

No evidence for a longer-lived At^{206} was observed. Since the counting rates obtained from the alpha particles of Po^{206} were always quite low, it was not possible to observe its growth. Thus an experiment was designed specifically to look for any long-lived At^{206} isomer. A stack of foils consisting of three electroplated platinum targets (enriched in Pt^{198}) was bombarded with N^{14} ions for approx 3 hr. At the end of bombardment, recoil catchers containing the astatine products were divided into two portions. The astatine fraction from one half of each recoil catcher was isolated approximately 20 min after the end of bombardment, and decay of the various astatine isotopes was followed in the alpha-particle pulse-height analyzer. About two hours later, astatine from the other halves of the catchers was separated and the samples pulse-height analyzed. Later, the alpha-particle radiations emitted by all the samples were counted for long periods of time in order to obtain good statistics on the amount of Po^{206} on each. The samples from the two halves of each recoil catcher were normalized to the same chemical yields by use of the amounts of At^{205} activity observed on each in the early, short counts. The results of this experiment make it possible to set the following upper limits on the ratios of 2.6-hr At^{206} /29.5-min At^{206} at the indicated N^{14} bombarding energies: 75 Mev, 0.04; 91.5 Mev, 0.022; 103 Mev, 0.031.

4. At^{204} - Po^{204}

In 1951, Barton, Ghiorso, and Perlman¹³³ reported the discovery of At^{204} . The half life, about 24 min, was determined by "milking" experiments in which polonium was extracted from the astatine at short intervals. Each of the polonium samples was allowed to decay for 5 hr before a

bismuth fraction was removed. From the amount of Bi^{204} present in the various samples, it was possible to determine the half life of At^{204} . No alpha decay was observed from this isotope, but, unless the alpha/EC branching is quite large, the alpha group from At^{204} would probably have been covered by a very intense group from At^{209} . The At^{204} atoms were formed in $\geq 150\text{-Mev He}^4 + \text{Bi}^{209}$ reactions.

Recently Hoff, Asaro, and Perlman have reported a 5.950-Mev alpha group,³² decaying with a half life of 9.3 ± 0.2 min, which they attribute to At^{204} . This activity was produced by the reaction $\text{Au}^{197}(\text{C}^{12}, \text{n})\text{At}^{204}$. They report no evidence for production of a 24-min isomer by this reaction.

In the experiments reported herein, it was not possible to separate the 5.95-Mev alpha particles of At^{204} from the intense 5.899-Mev group of At^{205} . However, semilogarithmic plots of the counting rate of the combined activities as a function of time could be resolved into one component of approx 26-min half life (At^{205}) and another of half life 9 ± 1 min, presumably the 9.3-min At^{204} of Hoff et al. The growth of the 5.37-Mev alpha particles from Po^{204} indicated a parent of 10 ± 5 -min half life. This figure could not be determined with much accuracy because of the small number of 5.37-Mev alpha counts observed during the short counts taken immediately after preparation of the astatine samples.

From the number of alpha particles emitted by the 9.3-min At^{204} and its daughter, Po^{204} , it was possible to calculate the percentage of alpha decay of At^{204} . In order to make this calculation, it was necessary to use the half life and alpha-branching ratio of Po^{204} . The half life has been found to be 212 ± 2 min. This is in good agreement with earlier determinations of 3.8 hr¹³⁸ and 3.5 hr.³⁸ The percentage decay by alpha emission has been estimated as about 1%,¹³⁸ and $4.2 \pm 0.4\%$.¹³⁹ Since both these values have been estimated from the yields of nuclear reactions and the expected cross sections, neither value is considered to be very accurate. Instead, a value of 0.63%, determined from the observed total half life and the partial alpha-decay half life, predicted from alpha-decay systematics, has been used.^{120,140} This analysis yields a value of

$4.4 \pm 0.2\%$ for the percentage decay by alpha-particle emission by At^{204} . This value is based on results of this study in addition to those of Latimer and Thomas.³⁵

The growth of Po^{204} alpha activity gave no indication of the presence of a 24-min isomer of At^{204} ; however, as noted above, the statistics were quite poor on the early counting rates of Po^{204} alpha particles. Because of this, an experiment was designed for more careful observation of a 24-min isomer. A 0.00025-in natural platinum foil was bombarded with nitrogen ions of 59 to 77 Mev energy. This was allowed to stand for about 45 min after the end of the bombardment in order that the 9.3-min At^{204} would have essentially completely decayed before separation of the astatine. The astatine fraction was then isolated from the polonium fraction by means of the double vaporization procedure discussed in Section III. The alpha particles emitted by the sample were observed in an alpha-particle pulse-height analyzer, and no growth of Po^{204} alpha activity was seen. From the amount of At^{207} present in the sample and the ratio of 9.3-min $\text{At}^{204}/\text{At}^{207}$ produced in other bombardments done under these conditions, it was possible to set an upper limit of 0.07 on the ratio of the yield of 24-min isomer to that of the 9.3-min isomer. This value is independent of the Po^{204} alpha-branching ratio.

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