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Emission of Long-Range Alpha Particles in the Fission of U^{238} with 17.5-MeV Protons*

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The angular distribution of long-range alpha particles from ternary fission induced by 17.5-MeV protons incident on U^{238} has been measured with respect to both the fission-fragment axis and the beam direction. For alpha particles emitted perpendicularly to the fragment direction, the distributions are found to be independent of the azimuthal angle about the fission-fragment separation axis and independent of the angle with respect to the incident beam. The angular distribution of ternary-fission fragments is nearly identical to that of binary-fission fragments resulting from the same initial system of fissioning nuclei. These results are compatible with a mechanism in which nuclei undergoing ternary fission are quite similar to those undergoing binary fission. A comparison was made of the angular correlation of long-range alpha particles and fission fragments for proton-induced fission of U^{238} and spontaneous fission of Cf^{252}. When no differentiation is made between light and heavy fragments, the full widths at one-half maximum are $47\pm12°$ and $33\pm2°$, respectively. Also the former system shows an unexpectedly large number of alpha particles emitted along the direction of the fragment separation axis. A ternary-to-binary fission ratio of $(2.7\pm0.6)\times10^{-3}$ for U^{238} plus 17.5-MeV protons was obtained from these measurements. The reported increase of the ternary-to-binary ratio at high excitation energies does not appear to depend in a simple way on the angular momentum of the initial compound nucleus. The binary-fission measurements yielded a fission cross section of 0.9 b and an angular anisotropy $[W(0)/W(90°)]$ of 1.12.

INTRODUCTION

ALTHOUGH the most common mode of fission is division into two fragments of roughly equal mass, in about one event in a few hundred a third light, charged particle is emitted. This phenomenon is often given the name ternary fission, although the same name is also sometimes used for the much rarer division into three more or less equal fragments.[1] Over 90% of the light, charged particles emitted in ternary fission are alpha particles, [2–4] and, hence, the terms "ternary fission" and "fission with long-range alpha particles" are frequently used interchangeably.

Two recent measurements related to the phenomenon of ternary fission have yielded rather unexpected results. In the first of these, Ramanna, Nair, and Kapoor,[5] induced fission in U^{238} with 14-MeV neutrons and measured the angular distribution of long-range alpha particles and their associated fission fragments with respect to the direction of the incident neutron beam. They observed that the alpha-particle distribution was peaked fore and aft along the beam direction and that the distribution of the associated fission fragments was peaked at 90° to the beam direction. These results are mutually consistent, since the alpha particle is emitted in a direction approximately perpendicular to the direction of the fission fragments.[6–9] They are, however, contrary to what one would expect from similar measurements on binary fission, where the fragment angular distribution is peaked fore and aft.[1] Furthermore, the ratio of differential cross sections for emission of alpha particles at 0° and 180° to that at 90° was reported to be extremely high, being about 10 to 1. These results seemed to indicate that fission with the emission of a long-range alpha particle is a different phenomenon from binary fission.

In the second experiment, Coleman, Fairhall, and Halpern [10,11] have measured the ratio of the ternary- to binary-fission cross sections at several different ex-

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[1] A review of what is known about the various phenomena known as ternary fission is given by E. K. Hyde, *The Nuclear Properties of the Heavy Elements* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1964), Vol. III, pp. 131–140.

[2] C. B. Fulmer and B. L. Cohen, Phys. Rev. **108**, 370 (1957).

[3] H. E. Wegner, Bull. Am. Phys. Soc. **6**, 307 (1961).

[4] J. C. Watson, Phys. Rev. **121**, 230 (1961).

[5] R. Ramanna, K. G. Nair, and S. S. Kapoor, Phys. Rev. **129**, 1350 (1963).

[6] E. W. Titterton, Nature **170**, 794 (1952).

[7] N. A. Perfilov and Z. I. Solov'eva, Zh. Eksperim. i Teor. Fiz. **37**, 1157 (1959) [English transl.: Soviet Phys.—JETP 10, 824 (1960)].

[8] M. L. Muga, H. R. Bowman, and S. G. Thompson, Phys. Rev. **121**, 270 (1961).

[9] Z. Fraenkel and S. G. Thompson, Phys. Rev. Letters **13**, 438 (1964).

[10] J. A. Coleman, A. W. Fairhall, and I. Halpern, Phys. Rev. **133**, B724 (1964).

[11] In a recent communication from A. W. Fairhall, we have been informed that the ternary-to-binary ratio for the alpha-particle-induced fission of U^{238} has been remeasured at the University of Washington. Rather than increasing at the higher excitation energies, the ternary-to-binary ratio is found to level off at a value of about 1×10^{-3}.

citation energies of the initial compound nuclei Pu^{242} and U^{236}. Their results together with those of others[12,13] are shown in Fig. 1. This ratio is seen to go through a minimum of about 10^{-3} at an excitation energy of 19 MeV. However, all of the data beyond the minimum are based on alpha-particle-induced fission. It is possible that the use of alpha particles as a projectile increases the probability for the observation of long-range alphas in coincidence with fission.[11]

A convenient system for a study of both of the phenomena described above is the Np^{239} compound nucleus formed by irradiation of U^{238} with 17.5-MeV protons. The nucleus so formed is close in Z and A to those formed either by neutron irradiation of U^{238} (U^{239}) or by alpha-particle irradiation of U^{238} and Th^{232} (Pu^{242} and U^{236}). The excitation energy of the original compound nucleus is 22.7 MeV, to be compared with 18.7 MeV for 14-MeV neutrons on U^{238} or 25 MeV for 30-MeV alpha particles on Th^{232} or U^{238}. The average angular momentum of the Np^{239} compound nucleus is calculated to be $3.7\hbar$, whereas that for the U^{239} nuclei formed in 14-MeV-neutron bombardment of U^{238} is $5.5\hbar$ and that for the Pu^{242} nuclei formed in 30-MeV-alpha-particle bombardment of U^{238} is $8.6\hbar$.[14–17]

We have made a series of measurements of the differential cross section for ternary fission induced by 17.5-MeV protons incident on U^{238}. Coincidences were required between an alpha-particle detector and a fission-fragment detector 90° apart. One detector could be fixed at a particular angle to the proton beam while the other was moved at various angles to the beam (consistent with the two detectors being 90° apart). In this way it was possible to measure a reasonably complete angular distribution of the long-range alpha particles and ternary-fission fragments with respect to the beam direction.

Coleman, Fairhall, and Halpern determined the ternary-to-binary ratio for each system they studied by comparing the coincident counting rate under the conditions of interest with the coincidence counting rate when a Cf^{252} source was placed in the same position as the target. They then assumed that the ratio of ternary fissions to binary fissions was equal to 3.3×10^{-3} times the ratio of the counting rate for the system of interest to the counting rate for Cf^{252}. (3.3×10^{-3} is the measured [13] ternary-to-binary ratio for Cf^{252}). However, it is possible that the angular correlation between the alpha particles and the fission fragments is different for the system being studied from what it is for Cf^{252}. We have therefore measured this angular correlation for both Np^{239} and for Cf^{252}.

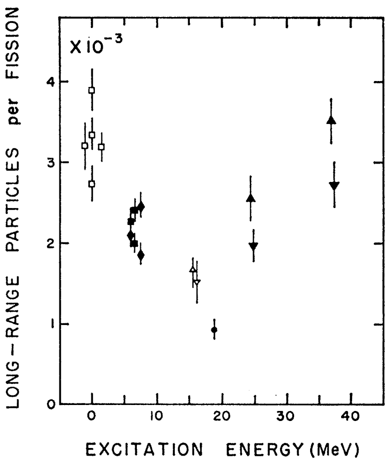

FIG. 1. The relative probability for ternary fission as a function of the initial excitation energy of the compound nucleus. This figure is taken from Ref. 10. Squares and diamonds refer to data of Nobles (Ref. 13) and the dot to a datum of Perfilov *et al.* (Ref. 12). Triangles refer to data of Coleman *et al.* (Ref. 10), open triangles signifying proton-induced fission and filled triangles alpha-particle-induced fission: However, see Ref. 11.

EXPERIMENTAL PROCEDURE

Source and Target Preparation

The coincidence measurements were made using uranium targets prepared by vacuum volatilization of uranium tetrafluoride from a hot filament onto nickel foils about 5 μ in. thick backed by 10^{-4}-in. copper for additional strength. The copper was subsequently removed by etching with a mixture of trichloracetic acid and ammonia. The thickness of these targets was from 200 to 400 μg/cm^2 of U^{238}. Each target was very uniform in thickness and contained little or no contaminant materials. During preliminary measurements the U^{238} (p,α) Pa^{235} reaction was briefly investigated using a target of a uranium oxide electrodeposited on 5-μin. nickel. The uranium tetrafluoride targets were not suitable for this purpose since the alpha-particle groups from the F$^{19}(p,\alpha)$O^{16} reaction obscured those from the same reaction on U^{238} at all except the extreme backward angles. However, the Q for the (p,α) reaction on O^{16} is about 10 MeV lower than the Q for this reaction on F^{19}, and the alpha-particle groups appear at correspondingly lower energies in the singles spectra. The uranium oxide targets were not of sufficiently good quality to be used for the coincidence measurements.

Two californium sources were used, one prepared by evaporation of a drop of solution containing the californium onto a thick platinum plate, which was sub-

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[12] N. A. Perfilov, Z. I. Solov'eva, and R. A. Filov, Zh. Eksperim. i Teor. Fiz. **41**, 11 (1961) [English transl.: Soviet Phys.—JETP **14**, 7 (1962)].

[13] R. A. Nobles, Phys. Rev. **126**, 1508 (1962).

[14] The average angular momenta of the various compound nuclei were calculated from transmission coefficients given by Huizenga and Igo (Ref. 15) for alpha particles and by Auerbach and Perey (Ref. 16) for neutrons. We used Auerbach's ABACUS-2 program (Ref. 17) and optical-model parameters given by Bate and Huizenga (Ref. 25) to calculate transmission coefficients and reaction cross sections for protons.

[15] J. Huizenga and G. I. Igo, Nucl. Physics **29**, 462 (1962); Argonne National Laboratory Report No. ANL-6373, 1961 (unpublished).

[16] E. H. Auerbach and F. G. J. Perey, Brookhaven National Laboratory Report No. BNL-765, 1962 (unpublished).

[17] E. H. Auerbach (unpublished).

sequently heated to red heat in order to destroy organic material. The other was electrodeposited from an ammonium-sulfate solution onto a nickel foil 5 μin. thick. Both sources were covered with nickel foils either 2.5 or 5 μin. thick in order to impede the self-transfer of the californium and contamination of the detectors and other equipment. These sources had an intensity of about 5×10^5 fissions per minute.

Detector Mounts

A detector mounting system was required such that measurements could be made at all angles with respect to the incident beam that were consistent with the two detectors being 90° apart. Conceptually, the simplest arrangement would be to have one detector located in the horizontal plane containing the incident beam and at an angle θ with respect to the beam. The other detector would then be somewhere on a circular ring centered at the beam spot with its plane perpendicular to the axis running between the beam spot and the first detector. The possible range of angles with respect to the beam for the second detector is from $\frac{1}{2}\pi-\theta$ to $\frac{1}{2}\pi+\theta$.

Such an arrangement has many practical disadvantages: It may require a much deeper scattering chamber than is available. It requires a target mount that allows the target to be rotated about both horizontal and vertical axes. It is redundant since each point on the ring above the horizontal plane is equivalent to some point below the plane. A practical improvement is obtained by using only the bottom half of the ring and rotating the whole assembly around the beam axis until both detectors are the same distance below the horizontal plane. If both are the same distance from the target they will then both be at the same dip angle below the plane. With such an arrangement, all angles of interest between the beam direction and the detectors can be obtained with dip angles between 0° and 45°.

The detector mounting system is shown in Fig. 2. The bar to which the mount was fastened could be externally rotated about a vertical axis through the center of the scattering chamber to any angle with respect to the beam direction. The detector could be lowered below the horizontal plane containing the incident beam along the arc of a circle having its center at the target beam spot. This dip angle could be varied from 0° to 45°. The other detector was mounted in the same manner.

Detectors

The detectors used were surface-barrier detectors obtained from ORTEC; all were circular in shape with an area of approximately 300 mm².

The one used to detect the long-range alpha particles had a nominal resistivity of 23 000 Ω cm. This detector was operated at a bias of about 40 V, which produced a depletion layer sufficient to stop 8-MeV protons and 32-MeV alpha particles. This thickness, just enough to stop the maximum-energy long-range alpha particles (∼30 MeV), was chosen in order to minimize the energy deposited in the detector by scattered protons. The detector surface was covered with a 0.001-in. aluminum foil to stop fission fragments and, in the measurements with Cf^{252}, the californium alpha particles.

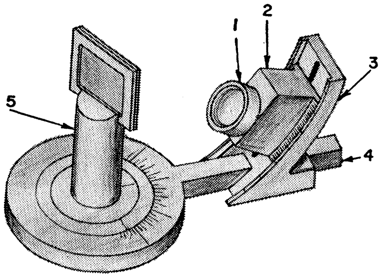

FIG. 2. A schematic drawing of the detector system. The solid-state detector (1) is attached to a block (2) which may be positioned anywhere along the curved surface of a mounting frame (3). The latter frame is attached to a bar (4) that can be rotated about the center of the target mount (5). An identical counter-mount system was attached to a rotatable plate just below the system shown. The apparatus shown was used for the Cf^{252} measurements; a similar, but not identical, system was used for the Np^{239} measurements.

The fission-fragment detectors were of a lower resistivity (150 to 1100 Ω cm) and were operated at a bias voltage sufficient to provide good charge collection. The leakage current through these detectors rose steadily during the course of the experiment because of the radiation damage produced by the fission fragments. It was therefore necessary to increase the applied bias voltage from time to time in order to compensate for the increased voltage drop in the load resistor. In this way it was possible to obtain reproducible fission-fragment spectra throughout the experiment.

Normally the detectors were located about 6 cm from the target and subtended an angle of about 16°. During the measurements of the angular distribution of binary-fission fragments two movable detectors were used to determine the fission counting rate at various angles with respect to the proton beam. These were located one on each side of the beam and about 18 cm from the target. In all of the experiments an additional fission detector, fixed at 45° with respect to the beam and about 25 cm from the target, was used to monitor the fission rate and, hence, the product of beam intensity times the number of target nuclei within the incident beam flux.

Electronics

Because the coincidence rate was extremely low (about 0.1 count/min) and because the beam intensity of the cyclotron could not be held constant, it was necessary to measure both the chance- and true-coincidence rates simultaneously. A convenient way to make such a measurement is to use a time-to-height converter to produce a pulse whose height is proportional to the time delay between two approximately coincident pulses. Events which are true coincidences correspond to a particular pulse height from the time-to-height converter; all other pulse heights represent chance events. If the output of the converter is analyzed in a multichannel pulse-height analyzer, the resulting spectrum is the desired delay curve.

It is possible to measure this delay curve as a function of the energy deposited in one of the detectors by analyzing the spectrum of pulses from the time-to-height converter on one axis of a two-parameter analyzer and the spectrum of pulses from one of the detectors on the other axis. With such a technique we were able to obtain, in a single experiment, the alpha-particle spectra for both true and chance events.

The charge from the solid-state detectors was converted to a voltage pulse in an ORTEC 103 integrating preamplifier. These pulses were further amplified in an ORTEC 203 amplifier operated in the delay-line mode. Each pulse was then fed to one panel of a Cosmic 801 coincidence circuit. The fast discriminators of these units were triggered by the zero crossing of the doubly differentiated pulses. These discriminators were used to generate the start and stop pulses for the time-to-height converter.[18]

The coincidence circuit also produced a gate pulse for the Nuclear Data 160 two-parameter analyzer if events were detected within a resolving time of about 300 nsec. The purpose of the gating pulse was to prevent the analyzer from accepting spurious pulses produced by the converter when a stop pulse happened to reach the converter just before a start pulse.

The output of the time-to-height converter was analyzed on one axis of the two-parameter analyzer and the output of the alpha-particle detection system on the other, thus giving a simultaneous recording of true and chance events. The stop pulse was delayed by about 160 nsec, so that a true coincidence produced a pulse in the middle of one axis of the analyzer. Thirty-two channels, each 10 nsec wide, were used for the delay curve; most of the true events fell into 4 channels. The ratio of the number of events in these channels to the average number of events per 4 background channels was about 5 to 1.

Cyclotron

The Princeton 20-MeV FM cyclotron was operated at 17.5 MeV. The external beam was focused by a pair of quadrupole magnets, deflected by a switching magnet, refocused, then momentum analyzed in a spectrometer magnet before passing into a 20-in. scattering chamber.[19] The beam intensity used was approximately 5×10^{-9} A with an energy spread of some 60 keV. In order to avoid pileup in the detector resulting from the large number of scattered protons and to keep the accidental coincidence rate down, the duty cycle of the external beam was increased by a factor of 10 using an auxiliary rf system to extract the beam.[20] Though the rf structure of the beam still persisted the beam extraction time was increased from 15 to 200 μsec. This increase in duty cycle coupled with the use of a time-to-pulse-height coincidence circuit enabled the experiment to be performed in spite of the difficulties normally encountered with coincidence experiments using beams from synchrocyclotrons.

RESULTS

Singles and Coincidence Spectra

A spectrum of reaction particles observed in the high-resistivity detector for 17.5-MeV protons incident on uranium tetrafluoride is shown in Fig. 3. The detector was covered with 0.001-in. aluminum foil to stop the fission fragments. The depletion depth of the detector was such that protons and alpha particles could deposit a maximum energy of 7.3 and 29.2 MeV, respectively, (corresponding to about 30-MeV alpha particles incident on the aluminum foil). The discrimination level was usually set at 8 MeV and the depletion depth increased until the maximum energy deposited by protons was just under 8 MeV. The peaks due to the Ni(p,α)Co reaction were identified by bombarding a thin nickel backing similar to that on which the uranium tetrafluoride was deposited; those due to the $F^{19}(p,\alpha)O^{16}$ reaction were identified similarly with a thin Teflon target. These peaks provided a convenient means of energy calibration. In Fig. 3 the arrow marked "LRA" shows where the center of the

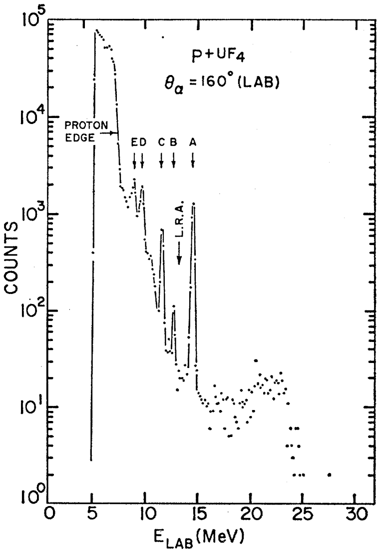

FIG. 3. A singles spectrum observed with the thick detector at 160° (lab) for 17.5-MeV protons incident on a uranium tetrafluoride target mounted on a 5-μin. nickel backing. The peaks indicated by letters were identified (see text) as resulting from the following reactions: *A*-F^{19}-$(p,\alpha_0)O^{16}$, ground state; *B*-$Ni^{60}(p,\alpha_0)$-Co^{57}, ground state; *C*-$Ni^{58}(p,\alpha_0)Co^{55}$, ground state; *D*-$F^{19}$-$(p,\alpha_1)O^{16*}$, 6.06–6.13-MeV doublet; *F*-F^{19}-$(p,\alpha_2)O^{16*}$, 6.93–7.13-MeV doublet.

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[18] D. L. Wieber, Nucl. Instr. Methods **24**, 269 (1963).

[19] A. Lieber, Nucl. Instr. Methods **26**, 51 (1964).

[20] H. O. Funsten, N. R. Roberson, A. Lieber, and R. Sherr, Rev. Sci. Instr. **35**, 1653 (1964).

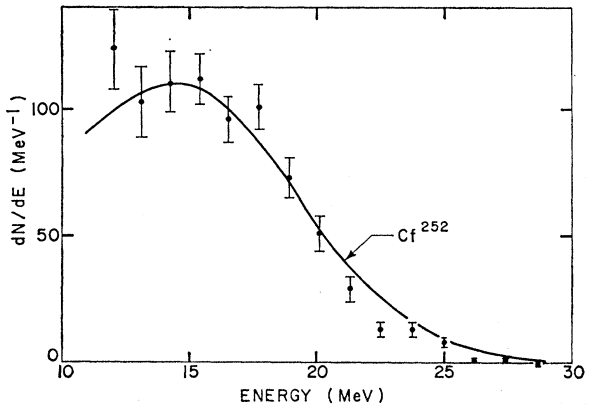


FIG. 4. The energy distribution of long-range alpha particles in coincidence with proton-induced fission of U^{238}. The errors shown are statistical counting errors. For comparison, the distribution of long-range alpha particles from spontaneous fission of Cf^{252} is indicated by the solid curve. Both distributions have been corrected for the distortion caused by the 0.001-in. aluminum foil covering the counter.

long-range-alpha-particle group accompanying fission should be located.

Alpha particles arising from the U$^{238}(p,\alpha)$Pa^{235} reaction should have energies ranging from about 18 MeV (the effective Coulomb-barrier height) to 27.5 MeV (the maximum energy possible for the given incident-proton energy). A broad peak covering this energy range is seen in the spectrum shown in Fig. 3 and is attributed to this reaction. This group was inspected at angles of 45°, 90°, and 163° using the uranium oxide target. The differential cross section associated with alpha particles in the higher energy portion of the spectrum (i.e., leading to low-lying excited states in Pa^{235}) was found to be somewhat forward peaked while that associated with alpha particles in the lower-energy portion was essentially isotropic. The total cross section for the (p,α) reaction on U^{238} was estimated to be on the order of 1 to 2 mb.

In practice, it would be difficult to distinguish between an event in which a long-range alpha particle originates during the proton-induced fission of U^{238} and one in which the alpha particle originates from the (p,α) reaction on U^{238} followed by fission of the residual nucleus Pa^{235}. However, the Pa^{235} nucleus must be left with enough excitation energy to exceed the fission threshold (about 6 MeV)[21] and for this to happen, the alpha particle must leave with less than 21 MeV of kinetic energy. Since the Coulomb barrier causes the lower end of the alpha-particle spectrum to cut off at about this energy, the latter process is not very probable. In addition, only a portion of these residual nuclei will undergo fission ($\Gamma_f/\Gamma_t=0.2$).[22] On the basis of our data and the above numbers, we have estimated that the (p,α) reaction followed by fission amounts to less than 1% of the coincident events that we observed.

Figure 4 shows the spectrum of long-range alpha particles in coincidence with fission of Np^{239}. This spectrum is composed of all the runs where the counters were 90° apart. The lower level cutoff was set so that the total true-to-chance ratio was sufficiently high to allow immediate inspection of the results of a run and make necessary modifications in beam intensity, etc. However, the major portion of the remaining chance events occurred in the bottom few energy intervals. In particular, a large chance rate in the lowest energy interval made it difficult to determine the position of the true-coincidence peak there, and for this reason, those data have not been used in the following results. For comparison, the solid line represents a singles spectrum of long-range alpha particles from Cf^{252} obtained in the high-resistivity detector covered with a 0.001-in. aluminum foil.

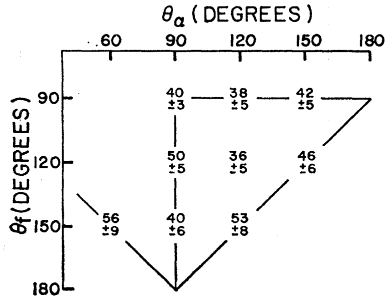
There is a likelihood that light, charged particles other than alpha particles contribute to the observed spectra. Protons are eliminated by setting the detector bias voltage so that protons deposit no more than 8 MeV in the detector and setting the discriminator level just above 8 MeV. On the basis of the work of Watson,[4] Wegner,[3] and Fulmer and Cohen,[2] we estimate that the contribution of other charged particles (mostly tritons) to the long-range-alpha-particle spectra is less than 3%. We can, therefore, refer to the particles detected as "alpha particles" with only a slight inaccuracy.

Angular Distributions

All possible combinations of the angles θ_f and θ_α that two detectors may make with respect to the beam can be represented by points in a two-dimensional space whose axes are θ_f and θ_α. The possible combinations in which the two detectors are 90° apart are confined to a diamond in this space. The quadrant of this diamond for both θ_f and θ_α greater than 90° is illustrated in Fig. 5. The numbers shown are the normalized, true-coincidence rates at the angles indicated. (All data are normalized to the monitor-detector counting rate.)

Within the accuracy of the approximation that the

FIG. 5. The angular dependence of the fission-alpha coincidence counting rate with the two detectors 90° apart. The rates are normalized to the monitor counting rate, and the errors are statistical counting errors.



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

[22] R. Vandenbosch and J. R. Huizenga, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy* (United Nations, Geneva, 1958), p. 284.

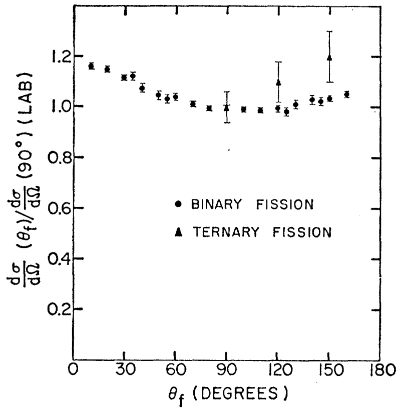

FIG. 6. The binary and ternary fission-fragment angular distributions (lab) with respect to the incident beam direction. Both distributions are normalized to unity at 90°.

directions of the two fission fragments are 180° apart, it would be redundant to make measurements with the fission counter at forward angles. A light fragment detected at an angle θ_l with respect to the beam is the partner of a heavy fragment at an angle $\theta_h = 180° - \theta_l$. Since no distinction was made between light and heavy fragments, the placing of a counter at some angle θ was equivalent to placing one at $180° - \theta$. The data are also expected to be symmetric about the $\theta_\alpha = 90°$ axis if the alpha particles are emitted in a compound-nucleus reaction or if the alpha-particle distribution is axially symmetric with respect to the fission-fragment direction. The forward- and backward-angle measurements at $\theta_f = 150°$ represent partial verification of the symmetry about the $\theta_\alpha = 90°$ axis.

To obtain the angular distribution of fissions in coincidence with long-range alpha particles, the data of Fig. 5 were averaged over all alpha-particle angles corresponding to a given angle of the fission detector. The results are shown in Fig. 6. These are plotted as a ratio to the 90° result since our measurements are only proportional to the cross section. For comparison, the laboratory angular distribution of binary-fission fragments is shown.

Similarly, for each angle of the alpha detector, the coincidence rates have been averaged over all corresponding angles of the fission detector to yield the long-range-alpha-particle angular distribution. These results are presented in Fig. 7. The solid curve shown is our representation of the data of Ramanna *et al.* normalized to unity at 90°.

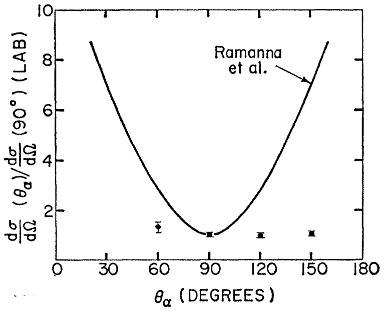

FIG. 7. The angular distribution (lab) of long-range alpha particles with respect to the incident beam for proton-induced (points) and neutron-induced (solid curve) fission of U^{238}. Both distributions are normalized to unity at 90°. The data on neutron-induced fission are from Ref. 5.

Angular Correlations

On the basis of previous results[6–9] it would be expected that to first order the distribution of alpha particles would be strongly peaked at 90° with respect to the fission fragments. In Fig. 8 we show the angular correlation between the alpha particles and fission fragments for both Np^{239} and Cf^{252}. Most of the measurements on Np^{239} were obtained with both counters at 90° with respect to the beam but at various appropriate azimuthal angles about the beam axis. However, some points were necessarily obtained with the counters at various angles in the horizontal plane. There are several features of interest in these angular correlations. Although the error bars are fairly large for the Np^{239} data, it appears that the correlation curve is somewhat broader than that for Cf^{252}. Further, it should be pointed out that the Np^{239} correlation function is weakly peaked again at 180°. Finally, it will be noticed that the correlation functions are not symmetric about 90° but rather about an angle several degrees larger than 90°. This effect is mostly a consequence of the fact that the fission fragments do not go exactly in opposite directions but are deflected from this angle by a total of about 5° when an energetic alpha particle is emitted. Hence an alpha particle emitted at 82° (90°−8°) with respect to the light fragment makes an angle of 103°(90°+8°+5°) with respect to the heavy fragment and not 98°(90°+8°).

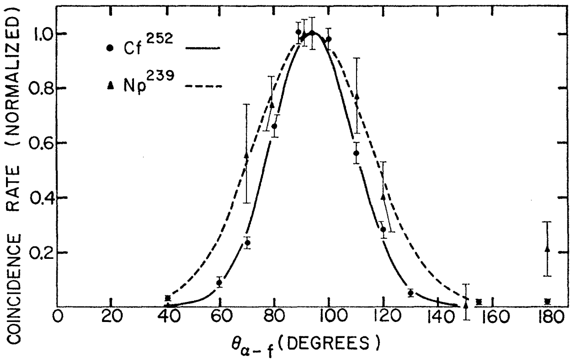

FIG. 8. The angular correlation between the fission fragments and long-range alpha particles for proton-induced fission of U^{238} and spontaneous fission of Cf^{252}. The curves represent the function $\exp[(\theta-\theta_0)^2/2\sigma^2]$ after folding in the experimental dispersion due to the detector's finite acceptance angles. The best fits to the data are obtained with $\theta_0 = 94°$ and $\sigma = 20°$ and 14° for Np^{239} and Cf^{252}, respectively. The errors are statistical counting errors.

The fact that there are essentially no true coincidences detected at an angle of 150° between the two detectors implies that there is no contribution from the $(p,\alpha f)$ reaction to the coincident rate. Such a strong correlation between the directions of the alpha particle and the fission fragment would not be expected for the $(p,\alpha f)$ reaction.

Ternary-to-Binary Ratio

Using the same technique as that used by Coleman *et al.*,[10] we have compared the coincidence counting rate for the Np^{239} system with that for Cf^{252} under identical experimental conditions. The ternary-to-binary ratio for the fissioning nucleus Np^{239} at an excitation energy of 22.7 MeV was taken to be 3.3×10^{-3} times the ratio of the coincidence counting rates of the respective systems. The value so obtained is $(2.1\pm0.2)\times10^{-3}$, in good agreement with the ratios reported by Coleman *et al.* at similar excitation energies.[11]

Implicit in this method is the assumption that the angular correlation between the long-range alpha particles and the fission fragments is the same for Cf^{252} and Np^{239}. Our results indicate that the Np^{239} distribution is broader than that for Cf^{252}. The above ratio, 2.1×10^{-3}, is therefore too low.

We have calculated a correction factor for this effect in the following way: We assume that the number of long-range alpha particles per unit solid angle is given by a Gaussian distribution centered about the measured angle of symmetry. This correlation function was integrated over the finite acceptance angles of the two detectors for each angular setting between them to give an angular-correlation function into which the experimental dispersion had been folded. The width of the correlation function was chosen by trial and error to give a resulting function that agreed with the experimental results. The curves giving the best fit are shown in Fig. 8. The values of the full width at half-maximum (FWHM) so determined are $33°\pm2°$ for Cf^{252} and $47°\pm12°$ for Np^{239}.

Using these widths we then calculated the geometrical factors for the two fissioning nuclei with the counters 90° apart. Multiplying the quantity 2.1×10^{-3} by the ratio of these geometrical factors we obtained a final answer for the ternary-to-binary ratio of $(2.7\pm0.6)\times10^{-3}$. Most of the quoted error arises from statistical error in the Cf^{252} measurement and uncertainty in the width of the Np^{239} angular correlation.

Binary Fission

If the angular distribution of binary fragments shown in Fig. 6 is transformed to the center-of-mass system the ratio of the differential cross section at 0° to that at 90° (the anisotropy) is found to be 1.12. We may compare this with the predictions of the Halpern and Strutinski[23] model. On the basis of ratios of neutron-emission width to fission width (Γ_n/Γ_f) given by Vandenbosch and Huizenga,[22] we estimate that 46% of the fissions occur before neutron evaporation, 34% after the emission of one neutron, and 19% after the emission of two neutrons. The third of these quantities is uncertain because the Np^{237} nucleus formed by the emission of two neutrons has an excitation energy very close to the fission threshold. Using the curve of K_0^2 versus excitation energy at the saddle point determined by Vandenbosch, Warhanek, and Huizenga,[24] we calculate an anisotropy of 1.07 for the combination of first- and second-chance fission. Any contribution of third-chance fission will tend to raise this number, but it is almost impossible either to estimate accurately how much third-chance fission takes place or to calculate the anisotropy for third-chance fission.

Our measurements give a cross section of 0.9 b for fission induced in U^{238} by 17.5-MeV protons. From similar measurements, Bate and Huizenga[25] have determined the reaction cross section for the interaction of protons with uranium nuclei over the proton energy range 4 to 12 MeV. From their results they have determined optical-model parameters consistent with their measurements. Using these same parameters we calculate a total reaction cross section at 17.5 MeV of 979 mb; combining this number with the values of Γ_n/Γ_f given by Vandenbosch and Huizenga we obtain a predicted fission cross section of 0.88 b in good agreement with the measured value.

CONCLUSIONS AND SUMMARY

Our results for the angular distribution of long-range alpha particles and associated fission fragments in 17.5-MeV-proton-induced fission of U^{238} are completely different from those reported for 14-MeV-neutron-induced fission of U^{238}. Within experimental error, we find that the angular distribution of alpha particles is isotropic and the distribution of fission fragments is the same as for binary fission. The results of this experiment are compatible with a model where the distribution of angular momenta in nuclei that undergo ternary fission is very similar to that in nuclei that undergo binary fission and in which the long-range alpha particles are emitted with azimuthal symmetry about the separation axis of the fission fragments. Of course, the system studied here is not identical to that studied in the neutron-induced fission experiment, the average angular momentum of the initial nuclei being 3.7 and $5.5\hbar$, respectively. However, it would seem to require a very selective mechanism for the few extra units of angular momentum to have such a large effect on the angular distributions with so little effect on the rate of production of long-range alpha particles.

Combining our measurement with those of others would lead to the conclusion that the number of long-range alpha particles per binary fision as a function of the excitation energy of the initial nucleus has a

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[23] I. Halpern and V. M. Strutinski, *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy* (United Nations, Geneva, 1958), p. 408.

[24] R. Vandenbosch, H. Warhanek, and J. R. Huizenga, Phys. Rev. **124**, 846 (1961).

[25] G. L. Bate and J. R. Huizenga, Phys. Rev. **133**, B1471 (1964).

minimum at about 19 MeV. The ratio for Np^{239} at an excitation energy of 22.7 MeV is significantly higher than those measured for similar systems at slightly lower energies. Our result is compatible with the large ratios found by Coleman *et al.*, but not with the more recent results of Fairhall.[11] Combining our result with previous measurements would imply that the variation in the ratio of ternary to binary fissions cannot be the result of some simple angular-momentum dependence; the initial compound nucleus Np^{239} has an average angular momentum which is less than that of the nuclei created in the work of Perfilov[12] on 14-MeV-neutron-induced fission and greater than those created in the work of Coleman *et al.*[10] on lower-energy-proton-induced fission but the ternary-to-binary ratio is higher than reported by either of these workers.

In our experiment the angular distribution of alpha particles with respect to the fission axis shows an unexpected small secondary peak along the fission axis. The particles detected at this angle are in about the same energy range as those detected at other angles. This fact rules out such possibilities as a fission-fission coincidence resulting from pinholes in the foil covering the alpha-particle detector since the average fission fragment has much greater energy than the highest energy alpha particle. Further, the foil was of generally good quality as evidenced by the absence of such 180° peaking in the Cf^{252} measurements. It should be recalled that our angular-correlation results are given in terms of coincidences per unit solid angle, whereas results obtained with emulsions are usually given in terms of events per unit polar angle. Any such effect would not stand out so prominently in data displayed in the latter manner.

In order to compare our alpha-fission angular correlations with those observed at other laboratories where the angle of the alpha-particle detector was measured with respect to the direction of the light fission fragment it is necessary to fold those results about an appropriate symmety axis to produce a curve corresponding to the result which would have been obtained if no discrimination was made between light and heavy fragments. Conservation of momentum applied to the fission process in which a long-range alpha particle is emitted yields an angle of about 92.5° for the symmetry axis. Fraenkel and Thompson[9] obtained a FWHM of 32° and a most probable angle of 81° with respect to the light fragment for the Cf^{252} alpha-fission angular correlation. Folding their data about 92.5° yields a FWHM of 49° compared to our measured value of $33°\pm2°$. The discrepancy may be partially resolved by noting that a measurement of the angular correlation with two fission detectors 180° apart yields a most probable angle of alpha-particle emission with respect to the light fragment which is too small. Making the simplest assumption, i.e., that the error is one-half of the amount that the fission-fragment directions deviate from being 180° apart, leads to a corrected value of 83.5° for the most probable angle. Applying this correction to the data of Fraenkel and Thompson, we calculate the width with only one fission detector to be 40°. Measurements by Perfilov and Solov'eva[7] of the alpha-fission angular correlation for thermal-neutron-induced fission of U^{235} have yielded a FWHM of about 25° and a most-probable angle of alpha-particle emission of 82° with respect to the light fission fragment. Folding their data about the symmetry axis gives a FWHM of 42°. This value is within the experimental error of the width ($47\pm12°$) measured for the Np^{239} system in our experiment.

Although some variation is to be expected in comparing angular-correlation measurements because of different values for the low-energy cutoff of the alpha-particle energy spectrum, the error due to this difference should not be more than a few degrees. Our result that the correlation function for Cf^{252} is narrower than that for Np^{239} is not unexpected. The mass and charge division are more symmetric in the former case and the total charge per fragment is greater. Other conditions being the same, the most probable angle of alpha-particle emission will be closer to the symmetry axis for Cf^{252} and the alpha particles will be more strongly focused at the most probable angle. Folding the distributions about the symmetry axis will then result in a narrower correlation function even when only one fission detector is used in the measurement.

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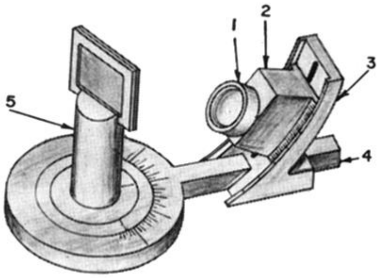

FIG. 2. A schematic drawing of the detector system. The solid-state detector (1) is attached to a block (2) which may be positioned anywhere along the curved surface of a mounting frame (3). The latter frame is attached to a bar (4) that can be rotated about the center of the target mount (5). An identical counter-mount system was attached to a rotatable plate just below the system shown. The apparatus shown was used for the Cf^{252} measurements; a similar, but not identical, system was used for the Np^{239} measurements.