Alpha-Particle Emission During Nuclear Fission at Moderate Excitation Energies*

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Protons, deuterons, and helium ions of various energies up to 42 MeV were used to induce fission in thin targets of Th^{232}, U^{235}, U^{238}, and Np^{237}. α-particle emission during fission was detected by a system of solid-state detectors coupled to a coincidence-gated pulse-height analysis system. α particles emitted prior to fission were distinguished from fission-accompanied α particles by the tight angular correlation between α-particle and fission fragments which prevails in the latter case. It was found that the probability of α emission during fission is insensitive to excitation energy over the energy interval from about 5 to 38 MeV for a variety of heavy nuclei from thorium to plutonium. There is a suggestion that α-particle emission is fractionally more probable for spontaneous fission than for fission of the same nuclei excited to energies ≥ 5 MeV. α-particle emission in the fission of At^{213} ($Bi^{209}+42$ MeV α) was detected, the probability of α-accompanied fission being about $\frac{1}{3}$ of that for a typical thorium or uranium nuclide. A possible effect of angular momentum on the probability of α emission during fission was looked for by forming Pu^{239} in two different ways, the excitation energy being the same but the angular momentum being different. No significant effect was observed. A correlation between the energy of the α particles and the sharpness of their angular distribution relative to the fission fragments, which had been seen at lower energies, was observed here to hold as well at higher excitation energies. In an experiment to determlne whether a correlation exists between the probability of α-particle emission and the mass ratio of the two heavy fission fragments, there is a suggestion that near-symmetric fragments have slightly less probability of emitting an α particle than asymmetric heavy fragments, although the effect is not very pronounced.

I. INTRODUCTION

THE study of the emission of α particles and other charged particles in coincidence with fission serves as a source of information about the nuclear configuration and dynamics at the instant of separation of the fission fragments.[1] The probability of charged-particle emission during fission is however very small. This has in the past discouraged investigation of these particles, especially at energies requiring the use of accelerator bombardments. Very recently however a number of new experimental results have appeared[2–8] which deal with various aspects of the problem of particle emission during fission.

The present paper reports some observations of α-particle emissions which occur in fissions produced in charged-particle bombardments of a few heavy elements. After a description of the experimental procedures (Sec. II) results concerning the dependence of the α-particle emission rate on a number of variables are considered. Section III A deals with the dependence of this rate on the excitation energy of the fissioning species, Sec. III B with the dependence on the Z and A values of the fissioning species, and finally Sec. III C discusses the dependence on the angular momentum of the fissioning nucleus. Section IV describes the results of measurements having to do with correlations involving α particles, Sec. IV A deals with a possible correlation of the α-particle emission rate with the fission-fragment mass distribution, and Sec. IV B deals with the correlation between the angular and energy distributions of the emitted α particles.

II. EXPERIMENTAL PROCEDURES

The fission targets (80 to 200 μg/cm^2 thick) were placed in the center of a large scattering chamber and bombarded with a collimated beam ($\sim$3 mm diameter). Proton bombardments were carried out at the University of Washington tandem accelerator and bombardments with α particles and deuterons were performed at the 60-in. cyclotron. Where it was necessary to lower the energy of the cyclotron beam, aluminum degraders were inserted into the beam collimator assembly.

Both the fission fragments and coincident charged particles were observed with semiconductor detectors. With one exception, to be mentioned later, the charged-particle detector was a $\Delta E-E$ telescope, the ΔE detector being a transmission-mounted surface-barrier detector with depletion depth of 250 μ. By requiring deposits of at least 11 MeV in the ΔE detector, accepted signals were limited to those produced by particles with $Z\geq 2$. (The largest amount of energy that singly charged particles could deposit in the ΔE counter was 8 MeV.)

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[1] I. Halpern, *Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965), Vol. II, p. 369.
[2] Z. Fraenkel and S. G. Thompson, Phys. Rev. Letters **13**, 438 (1964); Z. Fraenkel, Phys. Rev. (to be published).
[3] A. Gillet, T. P. Doan, C. Carlis, and R. Chastel, Compt. Rend. **262**, 296 (1966).
[4] T. D. Thomas and S. L. Whetstone, Jr., Phys. Rev. **144**, 1060 (1966); S. L. Whetstone, Jr., and T. D. Thomas, *ibid.* **154**, 1174 (1967).
[5] R. Ramanna, K. Nair, and S. Kapoor, Phys. Rev. **129**, 1350 (1963).
[6] H. Schmitt, J. Neiler, F. Walter, and A. Chetham-Strode, Phys. Rev. Letters **9**, 427 (1962).
[7] J. A. Coleman, A. W. Fairhall, and I. Halpern, Phys. Rev. **133**, B724 (1964).
[8] S. W. Cosper, J. Cerny, and R. C. Gatti, Phys. Rev. **154**, 1193 (1967).

The observed particles with $Z \geq 2$ presumably consist almost entirely of α particles. The total yield of other types of third particles with $Z \geq 2$ which have been observed in fission is less than 5% of the α-particle yield.[8] The particles detected in coincidence with fission by the biased charged-particle detector will therefore be referred to in what follows as α particles.

The largest amount of energy that an α particle can deposit in the ΔE counter is 23 MeV. Since the spectrum of α particles emitted in fission extends somewhat above 23 MeV, it was necessary to place an E counter behind the ΔE counter in order to avoid confusion in the upper portion of the spectrum. The 11-MeV bias in the ΔE counter cuts off a small portion of the lower end of the α-particle spectrum normally seen in fission. However, by determining the efficiency of the detecting system through a measurement of the (known) rate of α-particle-accompanied fissions in Cf^{252}, it was possible to make a first-order correction for the low-energy cutoff in the α-particle spectrum. That is to say, one can find the number of α particles per fission produced in a given sample, by comparing it to a reference source, even if the detecting system does not have 100% efficiency for all α-particle energies. This is because the spectra and angular distributions for fission α particles seem to be very similar for all species.[4] The energy calibration of the charged-particle detector was carried out with a combination of natural α emitters and the scattering of an α-particle beam from Au and C targets.

The fission-fragment counters were low-resistivity ($\sim 400\ \Omega$ cm) large-area (199 mm^2) surface-barrier detectors. The thin depletion depth ($\sim 60\ \mu$) permitted the observation of the full fission-fragment spectrum free from contaminations of beam particle pile-ups. The energy calibration of these detectors was carried out with the help of a thin Cf^{252} spontaneous fission source.[9]

After amplification, the fission-fragment and α-particle pulses ($E+\Delta E$) were sent to analog-to-digital converters. The outputs of these converters were printed out on paper tape when proper gating signals were received. The gating requirements were that the fission-fragment and α-particle pulses had to occur within 50 nsec of each other and, as already mentioned, that the ΔE pulse had to exceed 11 MeV.

Because of the low cross sections for fission accompanied by charged-particle emission, it was necessary to run at fairly high beam currents (30 to 200 nA) and to place the counters rather close ($\sim$1.3 in.) to the target. The determination of the effective solid angles subtended by the counters was part of the over-all efficiency determination mentioned above. For this purpose a thin Cf^{252} source was placed at the location of the beam spot and with the counter at 90° to the fission counter, the ratio of α-f coincidences to fission counts was determined. This ratio together with the known[10] rate of α's emitted per fission in Cf^{252} (3.35×10^{-3} α's per fission) permits one to convert the coincidence to singles ratios observed in bombardments to numbers of α particles emitted per fission. In making this conversion, one is assuming that α, f coincidences from Cf^{252} and those observed in reactions are being counted with the same efficiency. This may not be true because, as has been suggested,[4] the angular correlations between the α particles and fission fragments may not have the same widths in all cases. The accurate measurement of such widths is however difficult, largely because of effects of fission-fragment scattering. It was therefore decided not to attempt any corrections for possible differences in the widths of the α, f correlations between the targets used and the Cf^{252} standard source.

Because of the small cross sections it was necessary, in some cyclotron runs, to tolerate conditions where the accidental rates were almost half as large as the true rates. (The accidental rates in all tandem Van de Graaff runs were less than 1% of the true rates.) To determine accidental spectra, in the cyclotron runs, for subtraction from the observed spectra, the fast charged-particle signal was split. One signal was sent to the normal coincidence circuit and the other was delayed and sent to a second coincidence circuit. The delay was 87 nsec, i.e., one cyclotron period. The output of the second coincidence circuit was used to form a gate signal for accidentals. Pulse heights for both normal and accidental (i.e., delayed) events were recorded and distinguished by the printer. It should be mentioned here that the total amount of accidental events is not properly given with a system such as that described when succeeding beam bursts are of unequal intensity. Then the rate of overlaps of unrelated events, one from

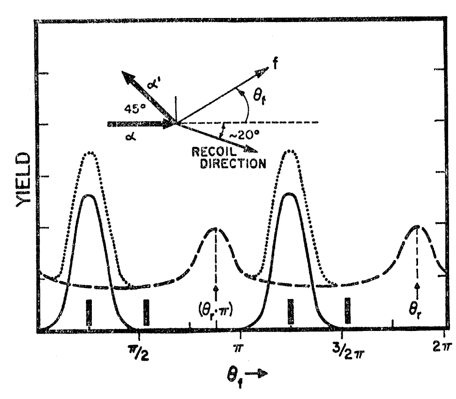

FIG. 1. The expected form of the α-fission fragment coincidence counting rate as a function of the angle of the fission detector. The fission detector is in the plane determined by the projectile and the emitted α particle. The α-particle counter is at 135° to the beam. The dashed curve gives the form of the counting rate for $(\alpha,\alpha' f)$ events and the solid curve for events in which the particles are emitted in conjunction with fission. The sum of the two curves (the dotted curve) is what one would observe.

[9] We are indebted to Dr. S. G. Thompson of the Lawrence Radiation Laboratory for supplying the Cf^{252} source used in these experiments.

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

each of two different beam bursts, is less than that of pairs of such events from the same beam bursts. Succeeding beam bursts in the cyclotron were found to vary greatly, and more or less randomly in intensity and it was necessary to use a separate circuit to determine a correction factor to apply to the measured number of accidental events.[11] This correction factor was generally of the order of 10%, and was easy to measure.

Although the foregoing paragraphs describe the experimental arrangement typically used in the various measurements, there were some variations. These will be described, where necessary, as each of the measurements is discussed in turn.

III. RESULTS ON THE DEPENDENCES OF THE α-PARTICLE EMISSION RATE

A. Dependence of the α-Particle Emission Rate upon Excitation Energy

In measuring the number of α particles emitted in the course of fission, it is important not to include those coincident events where the α particles are emitted just prior to fission. For example, in fission induced by α-particle bombardment one must be sure that events are not being counted where inelastic α scattering happens to give rise to an excited nucleus which fissions. In order to distinguish the distributions connected with prior α particles from those of interest in the present measurement, it is possible to make use of the strong 90° correlation of true fission α particles with the fragment direction. This technique had been used before[7] but (possibly due to poor statistics) it had been erroneously concluded that the rate of α-f coincidences which arise from inelastic α-particle scattering is negligible. Present measurements indicate that these events, which will be labeled $\alpha,\alpha'f$ events, may constitute as much as $\frac{1}{3}$ of the measured events of 42 MeV. Moreover the cross section for these extraneous events has a sufficiently strong dependence on bombarding energy to distort the trend of the dependence of the desired f,α events upon bombarding energy, if one fails to subtract the undesired events from the observations.

Figure 1 illustrates schematically the way in which a subtraction of $\alpha,\alpha'f$ events may be carried out. If an inelastically scattered α particle is observed in the α-particle counter, assumed fixed at 135°, then the nuclear recoil angle θ_R is roughly $-\frac{1}{2}(45°)$, the exact value depending on the energy of the inelastically scattered α particle. For the range of energies which can give rise to subsequent fissions, $\theta_R=-20°$ is a reasonable average value. The angular distribution of fission fragments which follow inelastic scattering and which lie in the α,α' plane is given by the dashed line in Fig. 1. This

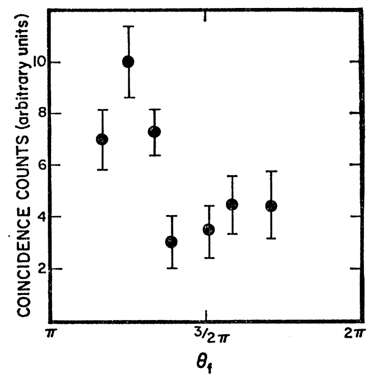

FIG. 2. The α-fission-fragment counting rate as a function of the angle of the fission detector in the bombardment of U^{238} with 42-MeV α particles. This curve has the form expected according to Fig. 1.

distribution is found to peak forward and backward along the recoil direction and to be symmetrical about the plane perpendicular to the recoil axis.[12] The solid line in Fig. 1 gives the distribution of fission fragments with respect to α particles emitted in the course of fission. The dotted curve, the sum of the two other curves, gives the distribution of the f,α coincidence rate that one would observe. From the aforementioned symmetries of the $\alpha,\alpha'f$ events it follows that the yields of these events must be the same at the four angles marked with black bars. Thus by subtracting the yield measured at the right-most angle having a black bar, $\theta_4=275°$, from that at $\theta_3=225°$ (the location of the third bar) one would presumably be left only with the true f,α events. This supposes that the angular distribution of the fission α particles is so narrow that it gives only a negligible contribution at θ_4.

A measurement with a target of U^{238} was made for the angular range $\theta_f=\pi$ to 2π with the α-particle detector at 135°, as in Fig. 1. The α-particle bombarding energy was 42 MeV and the results are shown in Fig. 2. It is seen that they are consistent with the expected pattern and that there is a relatively large contribution to f,α coincidences from inelastic scatterings followed by fission.[13]

About one-third of the events observed with 90° between the α particle and fission counters are seen to be $\alpha,\alpha'f$ events. Very similar ratios were observed when Th^{232} or U^{235} were bombarded with α particles of comparable energy. In a bombardment of bismuth, to

[11] I. Halpern, J. S. Heagney, D. L. Hendrie, and W. Loveland, University of Washington Cyclotron Progress Report. 1964, p. 45 (unpublished).

[12] B. Wilkins, J. P. Unik, and J. R. Huizenga, Phys. Letters 12, 243 (1964).

[13] It is possible to estimate the expected contribution of $\alpha,\alpha'f$ events from the measured spectra of α particles inelastically scattered at 135° from heavy elements [I. Halpern, J. S. Lilley, and N. Stein, University of Washington Cyclotron Progress Report, 1964, p. 13 (unpublished)] and from the known relative fission probability in excited uranium isotopes [R. Vandenbosch and J. R. Huizenga, in *Proceedings of Second United Nations International Conference on Peaceful Uses of Atomic Energy* (United Nations, Geneva, 1958), Vol. 15, p. 284]. The results of Fig. 2 are in reasonable accord with such an estimate.

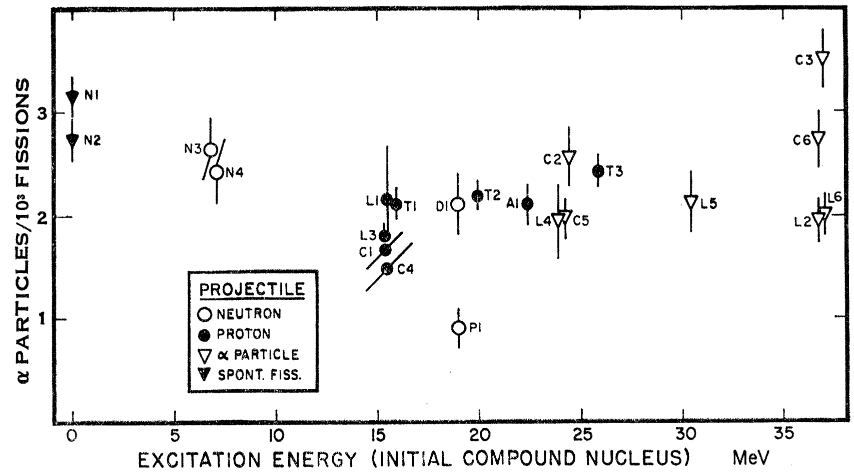

FIG. 3. The number of α particles emitted per fission observed by several authors in a number of different measurements. The data are plotted as a function of the excitation energy of the compound nucleus originally formed in the bombardment. The symbols defined in the inset denote the projectile and the letter symbols give the target and the author according to the following table:

L 1, 2	Th^{232}	Present work	*N* 1	Pu^{240}	Ref. 10
L 3,4, 5, 6	U^{238}	Present work	*N* 2	Pu^{242}	Ref. 10
A 1	U^{238}	Ref. 16	*N* 3	Pu^{241}	Ref. 10
C 1, 2, 3	U^{238}	Ref. 7	*N* 4	Pu^{239}	Ref. 10
C 4, 5, 6	Th^{232}	Ref. 7	*P* 1	U^{238}	Ref. 15
D 1	U^{238}	Ref. 17	*T* 1, 2, 3	U^{238}	Ref. 4

be discussed below, the ratio was much smaller. This is as one would expect since the range of excitations produced in α,α' on bismuth by 40-MeV α particles extends only to about 20 MeV. The probability for fission in bismuth does not begin to be appreciable until somewhat higher energies.[14] It was found in bombardments of Th and U isotopes by 21-MeV deuterons that the contribution from d,α events which are followed by fission was of a magnitude comparable to that found from $\alpha,\alpha'f$ events, namely about one-third of the observed events. In bombardments with 10.5-MeV protons, the background from direct p,α reactions which are followed by fission was found to be less than 1%. A negligible background must of course be expected in this situation on energetic grounds.

The total number of α-particle emissions per fission for a number of different bombardments was determined with the α and fission counters on opposite sides of the beam and at 135° to it. Where backgrounds due to fissions associated with α-producing direct reactions were large, these backgrounds were measured at the appropriate angles, as described above, and subtracted from the observations. The results are listed in Table I and are also plotted in Fig. 3 along with results of other authors.[4,7,10,15–17] The errors in the various measurements are typically ±15%. If one remembers that the points of Coleman *et al.*[7] for α induced fission at ~38 MeV (but not at ~25 MeV) are probably 30% too high because of the failure to remove the $\alpha,\alpha'f$ component, and if one also ignores the point of Perfilov *et al.* for 14 MeV neutrons,[15] it would appear that the number of α particles per fission has the nearly constant value 2.1×10^{-3} for all of the species in Fig. 3 for initial compound nuclear excitation energies above about 6 MeV. Each datum in Fig. 3 refers, of course, to the average value of α/f for a sequence of fissioning species beginning with the originally excited compound nucleus. The fact that these averages lie so close together for a variety of species over so broad a range of initial excitation energies suggests that α/f for each of the individual species in Fig. 3 is about 2×10^{-3} at all excitation energies above 6 MeV.

Thus α/f apparently has the following dependence on excitation energy. It falls somewhat as the energy varies from zero (spontaneous fission) to ~6 MeV (slow neu-

TABLE I. Probabilities for α-particle emission in fission.

Projectile	Projectile energy (MeV)	Target	Energy of initial compound nucleus (MeV)	Particles per fission	Symbol (Fig. 3)
p	10.5	U^{238}	15.4	$1.81\pm0.11\times10^{-3}$	*L*3
p	10.5	Th^{232}	15.5	2.15 ± 0.53	*L*1
α	29.0	U^{238}	24.0	1.94 ± 0.38	*L*4
α	35.5	U^{238}	30.5	2.13 ± 0.30	*L*5
α	42.0	U^{238}	37.0	2.01 ± 0.20	*L*6
α	42.0	Th^{232}	36.8	1.94 ± 0.22	*L*2

[14] J. R. Huizenga, R. Chaudhry, and R. Vandenbosch, Phys. Rev. **126**, 220 (1962).

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

[16] R. Atneosen, G. Garvey, and T. D. Thomas, Phys. Rev. **139**, B307 (1965).

[17] L. Drapchinskii, S. Kovalenko, K. Petrzhak, and I. Tyutyugin, At. Energ. (USSR) **16**, 144 (1964) [English transl.: Soviet At. Energy **19**, 69 (1965)].

tron fission) and then it changes more slowly, if at all, as the energy is further increased. The reason for this constancy may be connected with the observation that the average kinetic energy of fission fragments is independent of the initial excitation energy (see, for example, Ref. 20). This observation suggests that the excitation energy in excess of the saddle energy which is present in the nucleus remains as internal energy during the saddle to scission motion. It also suggests that the amount of such internal energy in the nucleus does not seriously influence the dynamics of the tearing apart in fission. More specifically, this may mean that the viscosity in the pulling-apart motion does not depend significantly on the internal excitation, for if it did, the rate of conversion of fragment kinetic energy to internal energy, as the nucleus moves from saddle to scission, would depend on the initial excitation energy. The observed mean fragment kinetic energy would then be expected to be dependent on the initial excitation energy. In short, it would seem from the constancy of the mean fragment kinetic energies that the tearing stage in fission proceeds more or less at the same rate at all excitation energies above the threshold. Since the probability of α-particle production in fission must depend very much on the dynamics of the tearing, the suggested stability in approach to scission may be the reason for the observed constancy of the α-particle production probability over a large range of initial excitation energies.[18]

The reason for the somewhat larger values of α/f in spontaneous fission than in higher-energy fission might be connected with the fact that in spontaneous fission the system does not start its motion down the far side of the barrier toward scission from the top of the barrier as it does in all other cases. It starts lower down and therefore moves somewhat more slowly at all stages. It is possible to construct models of the α-ejection process where α/f would benefit from such slower motion toward scission. However, at the present stage of our knowledge of the α-ejection process, all such models would be too speculative. The point of the present section was simply to call attention to a similarity between the dependences of the α/f ratio and the mean fragment kinetic energy upon initial excitation energy and to loosely explore implications of this connection.

B. The Dependence of the α-Particle Emission Rate upon Fissioning Species

The fact that the ordinates above ~6 MeV in Fig. 3 are very nearly the same for all of the species examined confirms earlier evidences of the weak dependence of α/f upon Z and A. Over the very limited range of species studied, the following empirical relation was found to be valid.[7]

$$\alpha/f=0.13(x-43)\times10^{-3}, \tag{1}$$

where $x=3.2Z-A$. According to this relation α/f increases for example from 1.7×10^{-3} for Th^{232} to 2.6×10^{-3} for E^{254}.

Although there exists no theory with which to compare this empirical description, it is easy to appreciate at least one factor that must play a role in the increase of α/f for heavier species. This factor is the increasing amount of distortion energy that becomes available at the scission stage. It is easy to estimate the average amount of excitation energy (distortion energy+internal nucleonic excitation energy) in the nucleus at scission. This energy is

$$E^*=E_R-E_K, \tag{2}$$

where E_R is the average (over-all fragments) of total energy release assuming that the nucleus is excited to its fission threshold energy. The quantity E_K is the average observed kinetic energy of the fragments. The mean distortion energy at scission E_D is necessarily smaller than E^*, but there is at present no good way to know what fraction of E^*, E_D happens to be. From mass tables,[19] average values of E_R for the two examples, Th^{232} and E^{254}, are 183 and 239 MeV, respectively. The average kinetic energies for the two species are 164 and 188 MeV, respectively.[20] Thus

$$E^*(Th^{232})=19\text{ MeV},$$
$$E^*(E^{254})=51\text{ MeV}.$$

These numbers give upper bounds on the mean distortion energy at scission. It is reasonable to assume[1] that it is the distortion energy at scission which is used to eject α and other particles. It has also been estimated[1] that the energy required for α-particle ejection, for example, is typically about 20 MeV. Thus the ejection of α particles is quite marginal in thorium. As is well known, the average fragment kinetic energy in α-accompanied fission is significantly smaller than the average kinetic energy in normal fission. This is interpreted to mean that only the events which reach abnormally large distortions achieve the chance to emit an α particle during fission. The value of E^* in E^{254} is not nearly so marginal as it is in thorium. Of course, without knowing about the division of E^* into E_D and internal energy, one cannot be sure that E_D for E^{254} is necessarily greater than it is for Th^{232}. But if even a small part of the extra E^* is reflected in increased values of the distortion energy for some of the events, this

[18] It is well to remember at this point that the probability of producing symmetric mass fragments depends very strongly on the initial excitation energy. For the details of mass division, the internal energy as well as the dynamics of separation are both apparently important. It is being suggested that for α-particle production on the other hand, the internal energy is not particularly relevant.

[19] J. C. D. Milton, University of California Report No. UCRL 9883 (Rev.), 1962 (unpublished).

[20] V. E. Viola, Jr., Nucl. Data **A1**, 391 (1966).

would help increase the rate of α-particle ejection. One may presume that the increase of α/f with the size of the fissioning nucleus arises in part from an increase in the probabilities of events with appreciable distortions.

These remarks about the heavy nuclei raise the question about α/f in lighter fissioning species (e.g., bismuth and radium). For the fission of bismuth with α particles, for example, the empirical relation (1) would suggest that $\alpha/f\simeq 1.25$, but to use the relation for bismuth involves its extrapolation far out of the range where it was found to hold approximately. In view of the small value of E^* for thorium, perhaps a more sensible way to guess α/f for a bombardment of bismuth is to see whether the trend of decreasing E^* for lighter species continues. If E^* (and hence E_D) becomes very small for bismuth fission, it will suggest that α emission should be very rare. If, however, E^* does not continue to decrease rapidly for lighter nuclei, it will mean that enough distortion energy for α-particle ejection may still be available. For At^{213} ($Bi^{209}+\alpha$) at the fission threshold, $E_R\simeq 164$ MeV and $E_K=152$. Thus E^* is 12 MeV. It does not fall off as rapidly between Th^{232} and At^{213} as it does between E^{254} and Th^{232}. On the basis of this value of 12 MeV one cannot completely exclude the possibility of α-particle ejection in the fission of At^{213}, especially when one takes into account the fact that less energy is needed for such ejection in At^{213} than in the heavy elements. On the other hand, one is certainly not encouraged to expect copious α-particle ejection.

A measurement of α/f in At^{213} in a bombardment of Bi^{209} with 42-MeV α particles gave the result (based on only 18 observed counts) that α/f in At^{213} is 0.3 ± 0.1 times what it is in a similar bombardment of uranium. In short,

$$(\alpha/f)_{\mathrm{At}}\simeq(0.6\pm 0.2)\times 10^{-3}.$$

The fact that the observed particles were fission α particles was verified by an examination of their energy and angular distribution. The energies of the 18 events lay between 11 and 18 MeV. In a search with an α-particle counter at 125° (rather than the usual 90°) to the fission fragments, no α-f coincidences were found in a time during which an average of 5 events would have shown up at 90°. Thus the α-particle distribution resembles the ones familiar from studies of heavier elements. The magnitude of α/f in astatine is apparently less than that in thorium, as one might have expected, but it is not an order of magnitude smaller.

C. Dependence of the α-Particle Emission Rate on the Angular Momentum of the Fissioning Nucleus

To see whether the ratio α/f was sensitive to the angular momentum of the fissioning species, the compound nucleus Pu^{239} was formed at a common excitation energy (30.3 MeV) in two different ways, (a) by bombardment of U^{235} with 35.5-MeV α particles, and (b) by bombardment of Np^{237} with 21-MeV deuterons. The values of the angular momenta of the compound nuclei which are formed in these bombardments are broadly distributed. The estimated rms initial angular momentum in the $\alpha+U^{235}$ bombardment ($12.1\hbar$) is significantly larger than that ($8.2\hbar$) in the deuteron bombardment. The measured ratio of α/f for these two bombardments was 0.92 measured to a statistical accuracy of 4%. If one allows for possible systematic uncertainties, this ratio is not significantly different from unity. Thus one must conclude that there is no strong influence of angular momentum on the α-particle emission rate in fission.

IV. CORRELATIONS OF THE α-PARTICLE EMISSIONS

A. Correlation between Fragment Mass Asymmetry and the Probability of α-Particle Emission

The mass distributions of α-accompanied fission have been found to be very similar to those for binary fission, when the fission is spontaneous or is induced by thermal neutrons.[2,6] That is, low-energy-fission studies show no particular connection between the probabilities of α-particle ejection and the fragment mass ratios. It is interesting to examine the same question in higher-energy fission because only at higher energies does the yield of nearly equal mass fragments (symmetric fission) become appreciable. There are evidences that symmetric divisions take place from larger fragment separations and therefore involve greater distortion energies at scission[21] than asymmetric divisions. Since the α-ejection rate presumably has to do with the distortion energy at scission, it is of interest to look for a possible correlation between mass-ratio and α-particle yield at energies where there is appreciable symmetric fission.

In order to study the correlation, a thinly backed target of U^{235} was bombarded with 13-MeV protons. At this bombarding energy there is essentially no α-particle emission before fission. In order to determine the fission-fragment mass ratio, two fission detectors were used, one on either side of the thin target. One of the counters subtended 20° with respect to the center of the target, and the other subtended 35°. This inequality in subtended angles is designed to ensure that the mates of all fragments which reach the counter of smaller solid angle will be recorded in the second counter. In computing the fragment mass ratio from the ratio of pulse heights observed in the two fragment counters, it is simplest to ignore recoil effects due to neutron emission. The effects are small. However if one ignores the larger recoil effects associated with the α-particle emission, it can be shown that one introduces an uncertainty in fragment masses of the order of five mass units. To reduce this uncertainty, one can take

[21] H. C. Britt, H. E. Wegner, and J. Gursky, Phys. Rev. **129**, 2239 (1963).

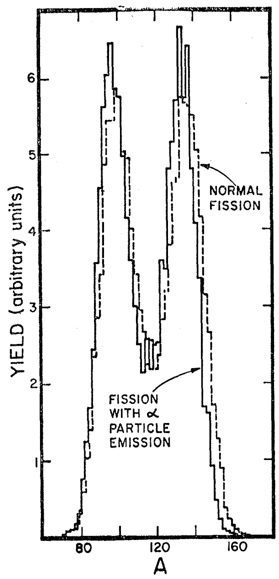

FIG. 4. The mass distributions in normal (binary) fission in the bombardment of U^{235} with protons and in those fissions in the same bombardment which are accompanied by α particles. The two curves have been normalized to the same total number of events. The errors associated with these two measurements are discussed in the text.

the recoil effect into account, to first order, by folding into the geometrical analysis, the measured[4] angular distribution of α particles with respect to the light fragment.

The α-particle detector used in the present measurement was for simplicity just the ΔE part of the charged-particle telescope which was used in the previously described measurements. The measured α-particle spectrum is distorted at its upper end when the ΔE counter is used alone. Because the errors introduced in this way are no larger than other errors which arise in the measurement, it was considered reasonable to use the counter as described. Simultaneous measurements were made with α-particle counters at 90° and at 75° to the axis of the fragment counters. The angles subtended by these two counters in the α-f plane were 45° and 30°, respectively.

The fragment mass spectrum seen in α-accompanied fission (where the α particle is observed in the 90° counter) is compared with the mass spectrum in normal binary fission in Fig. 4. It is seen that there is no striking evidence for increased symmetric yield in α-accompanied fission. To make the comparison between the two mass distributions somewhat more quantitative, the ratio of average yields for regions eight mass units wide on the peaks and in the valley were compared. The peak/valley ratio so defined was 2.29±0.09 for the binary fission and 2.50±0.12 for the α-accompanied fission. Thus, not only is there no evidence for more mass symmetry in α-accompanied fission, there may even be a slight suggestion that α-accompanied fissions are less symmetric than average fissions. It is important in this connection to remark that with the α-particle counter at 90°, the detecting system is somewhat more efficient for symmetric events (where the α particle tends to emerge perpendicular to the fragments) than it is for asymmetric events (where the α-particle angles are sometimes far off the perpendicular). These effects are fairly small because of the large solid angles used and were ignored. In any case, they would act in a direction that would enhance rather than diminish the symmetric yield. Because of these effects, events observed with the α-particular counter at 75° should have relatively more asymmetric mass divisions than those seen at 90°. This was indeed observed. The "peak/valley" ratio defined above was 3.02±0.26 at 75°.

Because of these preliminary yet provocative results regarding a possible correlation between the mass distribution and the probability for α-particle emission, it was considered desirable to investigate the same problem in the fission of radium, where the symmetric yield is more comparable to that of the asymmetric yield. Such measurements are now under way.[22]

B. The Correlation between the Energies and the Emission Angles of Emitted α Particles

Perfilov and Solov'eva[23] have reported that in the thermal neutron fission of U^{235}, the highest-energy α particles which are emitted in fission come off in the broadest angular distributions with respect to the fragments. The following results report the same kind of observation for higher-energy fission. Figures 5 and 6 show, for two bombardments, the coincidence counting rates as a function of the angle between the α-particle

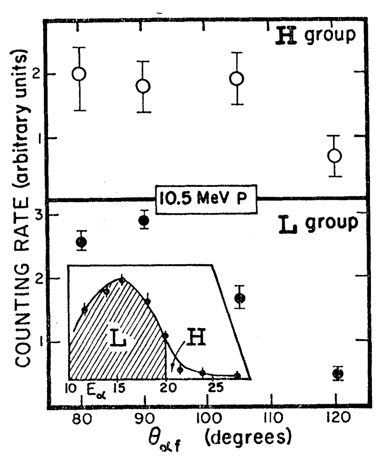

FIG. 5. Tne angular distribution of fission α particles with respect to the fission fragments in the bombardment of Th^{232} with 10.5-MeV protons. The α spectrum was divided into a low (L) and a high (H) energy group as shown in the inset.

[22] A. W. Fairhall, I. Halpern, W. D. Loveland, and D. G. Perry, University of Washington Nuclear Physics Laboratory Annual Report, 1966, p. 76 (unpublished).

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

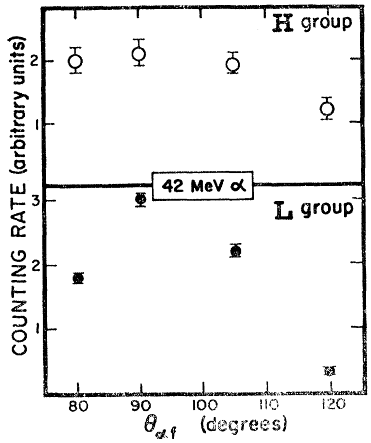

FIG. 6. The angular distribution of fission α particles with respect to the fission fragments in the bombardment of Th^{232} with 42-MeV α particles. H and L refer to the α particles above and below 20 MeV, respectively, as in Fig. 5.

detector and a fission detector. In these experiments the fission-fragment detector was placed at 135° and the α-particle detector moved relative to it at backward angles. In the α-particle bombardment this arrangement avoids confusing the fission α particles with inelastically scattered α particles. For purposes of examining the connection of these distributions with α-particle energy, the α spectra were divided into two groups. The group H refers to α particles having energies above 20 MeV and the group L to α particles having less than 20 MeV. (See inset Fig. 5.) It is seen that in both bombardments, (10.5-MeV protons and 42.0-MeV α particles in Th^{232}), the higher-energy α particles are more nearly isotropic than the lower-energy α particles.

It has been shown that the kind of correlation here exhibited points toward a limited range of fragment separations at the instant of scission. Were scission to occur at a great variety of fragment separations, those scissions which happen to occur when the fragments are still relatively close together would give rise to the most energetic α particles since the Coulomb potential at the place of α-particle release would still be large. These α particles would also come out sharply focused by the strong Coulomb field and would have a sharp angular distribution perpendicular to the fragment direction. On the other hand α particles which are released in scissions where the fragments are already well separated would tend to pick up less kinetic energy as they emerge from the now weaker Coulomb field and they would tend to be more broadly distributed in angle. In short, a broad distribution in separation distances at scission leads to a correlation opposite in character to that observed. These qualitative conclusions are supported by numerical calculations.[24] If the range of fragment separations at scission is small, the observed correlation can be understood in the following terms. The α particles which are released with greater initial energy tend to be less effectively focused by the Coulomb fields. They are therefore more broadly distributed in angle. They also tend to emerge with considerably more energy because they move out of the repulsive field before it is weakened by the motion of the separating fragments.

V. SUMMARY

The probabilities of α-particle emission and the angular distributions of the emitted α particles relative to the two heavy fragments are found to be remarkably similar in several bombardments of heavy-element targets. They also resemble the probabilities and distributions observed in spontaneous and slow neutron-induced fission. Only in spontaneous fission is there a suggestion that the probability of α-particle emission is slightly enhanced. The insensitivity of the α-particle emission rate to excitation energy is interpreted to mean that the scission stage proceeds in much the same way, independent of the internal energy of the fissioning system. The insensitivity of the fission-fragment kinetic-energy release is similarly interpreted to mean that the nuclear shape at scission is not very sensitive to internal energy.

There is a slight suggestion that α-particle emission may be more probable when the mass division in fission is asymmetric. The measurements also confirmed the correlation between α-particle energies and angular distributions which had been observed in lower-energy fission. Higher-energy α particles are emitted with a broader angular distribution with respect to the two heavy fission fragments than lower-energy α particles.

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[24] I. Halpern, CERN Report No. CERN-6812, 1963 (unpublished).