

Long-Range Particle Emission in Coincidence with Fission at Moderate Excitation Energies

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The production rate of long-range particles in coincidence with fission has been studied in bombardments of Th^{232} and U^{238} with 10.5-MeV protons and with helium ions of 29.5 and 42.0 MeV. The number of coincident charged particles per fission observed in the proton bombardments is consistent with the numbers of such particles observed in earlier measurements (spontaneous fission, neutron-induced fission). These results indicate that the production rate of charged particles in fission decreases smoothly with increasing excitation energy of the fissioning compound nucleus. For a nucleus excited to 15 MeV, the rate is about half that for a nucleus in its ground state. The charged particle yields obtained in the helium ion bombardments are, however, much larger than one would expect on the basis of the excitation energy dependence indicated by the other (lower energy) measurements. Some possible explanations for these observations are briefly and qualitatively considered.

I. INTRODUCTION

CHARGED particles observed in coincidence with fission are found to be emitted in a direction very nearly perpendicular to the motion of the fission fragments.¹ This observation has very reasonably been interpreted to indicate that the charged particles accompanying fission are released in the space between the freshly formed fission fragments with an initial kinetic energy which is small compared to their Coulomb potential energy.² In this view, the emergence of these particles at right angles to the direction of the fragments arises simply from the effects of the strongly focusing Coulomb field which is provided by the fragments. The observed energy spectrum of the charged particles (almost exclusively alpha particles^{3,4}) is consistent with this view of their release. In other words, the alpha particles that appear in fission are released either during or just after that final stage in the fission process, the so-called scission stage, when the nucleus actually divides into two separated fragments. The details of the production of these α particles are therefore particularly worth studying because they very likely can provide unique information about the final stages in nuclear fission.

Unfortunately, the α particles are emitted in less than one percent of the fissions and this low production rate has heretofore limited their extensive investigation. However, recent technical developments, such as that

of the solid-state detector, have led to a revival of interest and activity in this field.

From earlier work it appears that the number of α emissions per fission decreases as the excitation energy increases. In spontaneous fission, it has been found that an α particle is emitted in one out of every three or four hundred fissions.^{1,5} There are a number of measurements, the most extensive being those of Nobles,⁶ which indicate that in fission induced by thermal neutrons, the α -particle emission rate is about 25% lower. Recently Perfilov, Solov'eva, and Filov⁷ have reported that in fission induced in U^{238} with 14-MeV neutrons, the number of α particles per fission is lower still, being but one α particle in 1050 ± 100 fissions. These authors suggest, in fact, that their observation is consistent with the assumption that only those fissions following neutron emission can be accompanied by α particles. This would mean that when the excitation energy of the fissioning nucleus exceeds 10 MeV or so, no α particles are emitted in fission. It will be seen (Sec. III) that the more recent measurements lend some qualitative support to this view. The excitation energy dependence seems, however, to be somewhat weaker than Perfilov *et al.* suggest.

It also is known from earlier work⁶ that the α -particle production rate depends upon the fissioning species as well as on the excitation energy. The larger the atomic number of the fissioning species, the greater the number of α particles emitted per fission.

The present investigation provides new information

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¹ See, for example, N. A. Perfilov, Yu. F. Romanov, and Z. I. Solov'eva, *Usp. Fiz. Nauk* **71**, 471 (1960) [translation: *Soviet Phys.—Usp.* **3**, 542 (1961)].

² Tsien San-Tsiang, *J. Phys. Radium* **9**, 6 (1948).

³ C. B. Fulmer and B. L. Cohen, *Phys. Rev.* **108**, 370 (1957).

⁴ H. E. Wegner, *Bull. Am. Phys. Soc.* **6**, 307 (1961).

⁵ G. W. Farwell, E. Segre, and C. Wiegand, *Phys. Rev.* **71**, 327 (1947); Tsien San-Tsiang, Ho Zah-Wei, R. Chastel, and L. Vigneron, *J. Phys. Radium* **8**, 165, 200 (1947).

⁶ R. A. Nobles, *Phys. Rev.* **126**, 1508 (1962).

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

concerning the dependence of the number of α particles produced per fission (α/f) upon the nature of the fissioning species and especially upon the excitation energy. The range of excitation energies investigated here extends beyond that which has been explored so far. It is found that although the values of (α/f) observed in the bombardment of Th^{232} and U^{238} with 10.5-MeV protons are consistent with all of the above-mentioned earlier investigations, in the bombardments leading to higher excitation energies they are not. When Th^{232} and U^{238} are bombarded with helium ions of 29.5 and 42.0 MeV, the observed ratio (α/f) is much larger than that expected from an extrapolation of the lower energy results. The results of the present measurements, together with some possible interpretations, are discussed in the third section of this paper. The second section is devoted to a description of the method of measurement and the apparatus used.

II. EXPERIMENTAL PROCEDURES

Thin targets of natural uranium (essentially U^{238}) and thorium (Th^{232}) were bombarded with helium ions and protons from the University of Washington cyclotron. Coincident events were observed with a pair of silicon surface-barrier semiconductor counters. One of these counters detected one of the two fission fragments. The other counter (covered with a 6.2-mg/cm² aluminum foil to stop all fission fragments) detected charged particles in coincidence with fission. The geometrical arrangement of the counters and target is illustrated in Fig. 1. In order to ensure reasonable coincidence counting rates, it was necessary to use counters with a large sensitive area (roughly $\frac{1}{2} \times \frac{1}{2}$ in.) and to place the counters close to the target (at 1.25 in.). These dimensions determined the angular resolution of the measurement. The size of the collimated beam spot on the target was relatively small ($\frac{1}{8}$ -in. diameter).

The targets consisted of uranium or thorium oxide on a thin VYNS⁸ backing. The oxide particles were selected by differential sedimentation from an aqueous slurry in order to limit their size. The targets were everywhere thinner than 0.5 mg/cm².

Bombardments were carried out with protons of 10.5 ± 0.1 MeV⁹ and with α particles of 42.0 ± 0.3 and 29.5 ± 1 MeV. The latter beam energy was achieved by degrading the cyclotron beam in aluminum foils.

The electronic circuitry was a typical fast-slow coincidence system. A fast coincidence was formed between the pulses from the two detectors in a resolving time equal to about half a cyclotron period (87 nsec). The output of the fast coincidence circuits and the outputs of a pair of discriminators acting on the slow outputs from the detectors were then placed in slow coincidence. The output of the slow coincidence circuit was

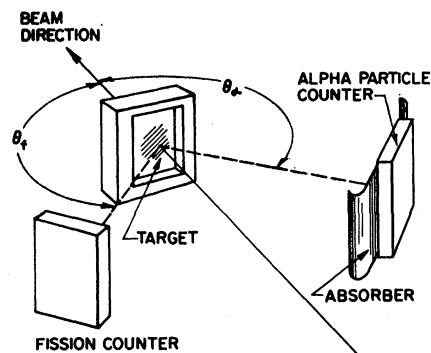


FIG. 1. The arrangement of target and (semiconductor) detectors in the experiment. The angles θ_f and θ_α were both 135° for most of the measurements.

used to gate a multichannel analyzer where the coincident fission fragment and α -particle spectra were simultaneously recorded side by side. It was possible to set the discriminator bias for the fission counter at such a level that essentially no fission pulses were rejected; but because of the large number of low-energy charged particles which emerged from the target, it was necessary to set the bias in the α -particle counter rather high in order to avoid accidental counts. The bias setting was determined with the aluminum absorber in place (Fig. 1) and corresponded to an α -particle energy, before the absorber, of about 10 MeV. Only about 10% of fission α particles are found to have energies less than this,^{6,10} and since the present measurement is relative rather than absolute (see below), it was felt that no corrections were needed for missed low-energy α particles.

The fission detector was thick enough to stop all fission fragments and the α -particle detector was thick enough to stop α particles up to well above 40 MeV. The pulse-height energy calibration for the α counter (determined by elastic scattering measurements and radioactive α sources of known energy) was linear to the highest energy (32 MeV) to which it was measured. There are few, if any, fission α particles with energies in excess of 30 MeV.^{3,6} Also, this calibration of the α -particle detector assured that no high-energy α particles (40 MeV) would appear as low-energy alphas. The fission detector was provided with a rough energy calibration simply by comparing its pulse-height distribution for fission fragments from the spontaneous fission of Cf^{252} with earlier measurements.¹¹ The fission spectrum from Cf^{252} was measured with the beam off and then also under typical operating conditions, with the beam on, in order to see if there were any pulse-height shifts arising from possible saturation effects in the

⁸ B. D. Pate and L. Yaffe, *Can. J. Chem.* **33**, 15 (1955).

⁹ I. Naqib, thesis, University of Washington, Seattle, 1962 (unpublished).

¹⁰ M. L. Muga, H. R. Bowman, and S. G. Thompson, *Phys. Rev.* **121**, 270 (1961).

¹¹ E. K. Hyde, University of California Radiation Laboratory Report No. 9036, 1960 (unpublished).

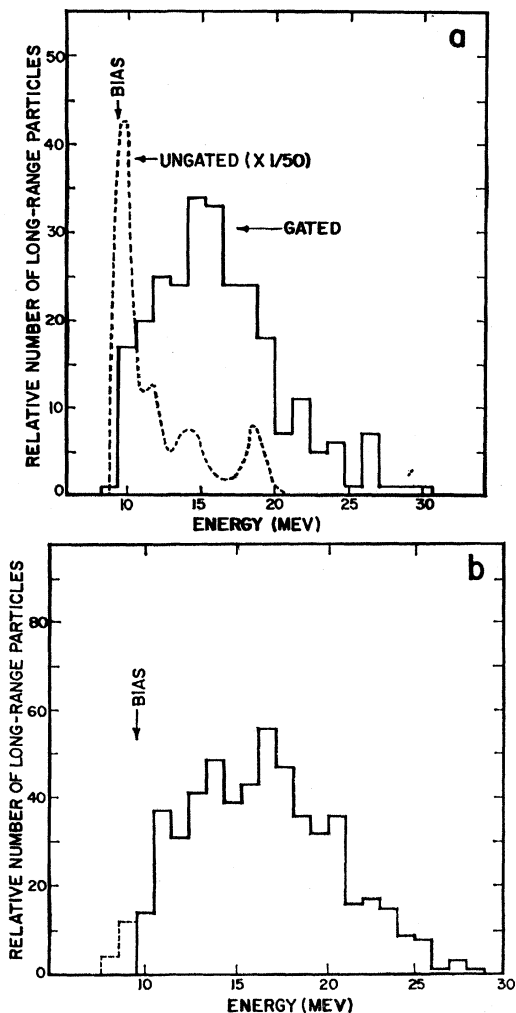


FIG. 2. The energy distribution of charged particles (assumed to be α particles) from fission (a) in the bombardment of Th^{232} with 42-MeV helium ions; (b) in the spontaneous fission of Cf^{252} . The thorium observations were made at 135° to the beam. The gated spectrum was obtained when a coincidence was required with a fission counter at 135° on the other side of the beam.

electronic equipment. The shift, if any, was less than 3% which is altogether negligible in the present experiment.

The ratio of true to chance coincidence rates during a run was typically about 10. To achieve these rates it was necessary to keep the beam currents below 4 μA for helium ions and below 20 μA for protons, and to keep the detectors in the backward hemisphere as described in the next paragraph. The chance coincidence rate was established by delaying the fast signal from either detector by a full period of the cyclotron. The insignificance of chance coincidence is indicated by the great difference between the coincidence spectrum in the α -particle counter and the ungated (singles) spectrum in the same counter [Fig. 2(a)].

Because of the copious yields of charged particles emerging from the target at forward angles, it was

possible to gather data efficiently only at backward angles. Accordingly, most of the measurements were made with the fission and α -particle counters at 135° on opposite sides of the beam. In order to convert the measured ratio of (long-range α -fission coincidences): (total fission counts in the same time interval) into a more useful number, the ratio was divided by the same ratio measured for a Cf^{252} spontaneous fission source under identical conditions. The result was then multiplied by the known number of α particles per fission for Cf^{252} , 3.35×10^{-3} .⁶ It is to be emphasized, then, that the present measurement is not an absolute measurement but is a *relative* measurement made under somewhat special conditions.

Despite the relative character of the actual measurements, it is reasonable to expect that the ratios (α/f) determined by comparison with Cf^{252} are close to those which one would obtain in measurements which integrate over all angles for both counters. The reasons for this expectation are based on the following observations:

1. The total yield of $\alpha-f$ coincidences was measured in Cf^{252} with the present apparatus and it agreed with the previously measured value within 20%. A measurement of the shape of the angular distribution in Cf^{252} was also consistent with the known shape¹⁰ when the (relatively poor) angular resolution of the present setup was taken into account.

2. The $\alpha-f$ angular distributions in the particle bombardments (e.g., $\text{Th}^{232}(\alpha, f)$ at 42 MeV) were found to be similar in shape to that in the spontaneous fission of Cf^{252} .

3. The fission counter orientation (135°) with respect to the beam was such that the observed fissions came from nuclei whose spin was close to the average spin of all of the compound nuclei produced in the bombardment.¹² Under these circumstances, it is reasonable to assume that the rate, (α/f), observed at 135° is close to the average (α/f) rate over all angles, even if this rate should happen to be spin-dependent.

Before turning to an account of the results, it is necessary to bring up some points concerning the identity of the particles observed in the α counter. Since only a single pulse height is measured in that counter, it is impossible to determine from the measurement alone what the mass of the observed coincident particles is; yet it seems that it can safely be presumed that the observed particles are α particles.

1. In other measurements, it is found that more than 95% of the charged particles emitted in fission are α particles.⁴

2. The observed pulse-height spectrum interpreted on the assumption that they are α particles, corresponds very closely to spectra of long-range α particles from

¹² I. Halpern and V. M. Strutinski, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958).

fission previously measured.³ Indeed, it would have been impossible for protons to make pulses in the α counter higher than those corresponding to an energy of about 9 MeV¹³ whereas the observed spectra generally extend up to 25 MeV or more.

In those bombardments which are performed with α particles, it is reasonable to wonder whether the α particles which are observed in coincidence with fission are truly scission α particles or whether they are, at least in part, inelastically scattered α particles which happen to induce fission. Even though inelastic scattering cross sections are small, so unfortunately are cross sections for fission with the production of α particles. One must therefore explore the extent to which scattering events may be contributing to the observations. The evidence that their contribution is insignificant comes from considerations of the energy and angular distributions of the observed particles.

1. *Energy distributions.* The observed coincident α -particle spectra [Fig. 2(a)] resemble those generally found in spontaneous and neutron-induced fission [Fig. 2(b)]. The expected alpha-particle spectra from inelastic scatterings followed by fission would be very different from the observed ones, peaking toward their high-energy end.

The observed coincident fission fragment spectra resemble the noncoincident spectra [Fig. 3(b)] rather than the double humped spectra which are characteristic of low excitation energy fission. The fissioning nuclei produced in inelastic scatterings would be expected to have fairly low excitation energy.

It should be pointed out here that the shift of the coincident fragment spectrum to lower energy than the noncoincident spectrum [Fig. 3(b)] is not by itself evidence for the nonscattering character of the events. Although such a shift is characteristic of alpha-particle accompanied fission¹⁴ [see Fig. 3(a)], it also would occur for fissions following inelastic scatterings. Such fissions would be occurring in nuclei where Z is two units less than in the compound nuclei which are produced in the bombardment. It has been observed,^{15,16} that the average kinetic energy released in fission goes roughly as $Z^2/A^{1/2}$ of the fissioning species with no strong dependence on excitation energy. This leads to an expected downward shift of the fragment spectrum for fissions following scatterings which is of comparable magnitude to the shift in genuine fission-alpha events.

2. *Angular correlations.* It was found that when either the fission or alpha-particle counter was moved to 90° to the beam (thus increasing the angle between counters from 90 to 135°), the coincidence counting rate was reduced by at least a factor of 2. It was even further re-

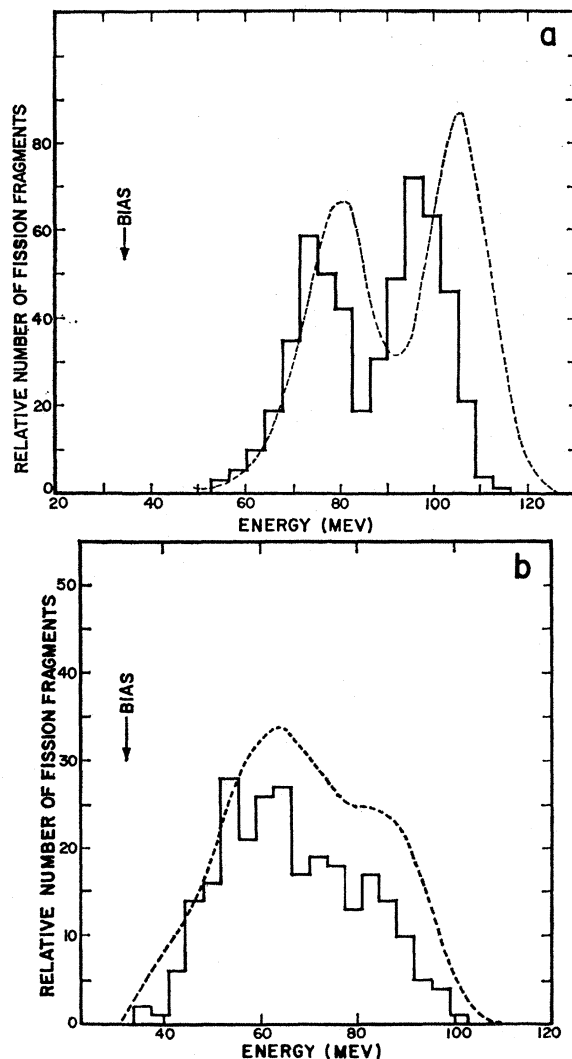


Fig. 3. The energy distributions of fission fragments (a) Cf^{252} ; (b) $\text{Th}^{232}+42\text{-MeV}$ α particles. The dashed curves were obtained with no coincidence requirement. The solid curves were obtained when a coincidence was required with a charged particle counter placed at 90° to the fission fragment direction.

duced when the counter was moved beyond 90° into the forward hemisphere. Since inelastically scattered alpha particles tend to go into the forward hemisphere, this dependence is not what one would expect for $(\alpha, \alpha'f)$ events, but it is exactly what one expects for genuine $\alpha-f$ events where the alpha particles show such a strong tendency to emerge at right angles to the fragments.

To sum up the last few paragraphs, the evidence from the shape of the observed α -particle spectra and coincident fission fragment spectra and from a rough exploration of the $\alpha-f$ angular correlation indicates that the observed α particles are scission α particles. There is no evidence for a significant contamination of the results in the α -particle bombardments by events associated with inelastic scattering.

¹³ G. Dearnaley, IRE Trans. Nucl. Sci. NS-8, 11 (1961).

¹⁴ V. N. Dmitriev, L. V. Drapchinskii, K. A. Petrzhak, and Yu. F. Romanov, Zh. Eksperim. i Teor. Fiz. 39, 556 (1960) [translation: Soviet Phys.—JETP 12, 390 (1961)].

¹⁵ I. Halpern, Ann. Rev. Nucl. Sci. 9, 245 (1959).

¹⁶ J. Terrell, Phys. Rev. 113, 527 (1959).

TABLE I. Probability for coincident α -particle emission in fission produced by charged particle bombardments of Th^{232} and U^{238} .

Incident particle and bombarding energy	Target	Compound nucleus	Initial excitation energy (MeV)	Number of observed events	$(\alpha/f)^a$
10.5 MeV p	Th^{232}	Pa^{233}	15.9	68	$(1.50 \pm 0.27) \times 10^{-3}$
	U^{238}	Np^{239}	15.8	155	(1.66 ± 0.20)
29.5 MeV α	Th^{232}	U^{236}	24.9	137	(1.96 ± 0.20)
	U^{238}	Pu^{242}	24.5	170	(2.56 ± 0.29)
42.0 MeV α	Th^{232}	U^{236}	37.4	531	(2.72 ± 0.28)
	U^{238}	Pu^{242}	37.0	1342	(3.52 ± 0.28)

^a The number of α particles per fission was determined from measurements taken with the α particle and fission counters at 135° on opposite sides of the beam. The over-all (α/f) ratios were deduced from a comparison with a standard Cf^{252} source as described in Sec. II.

III. RESULTS AND DISCUSSION

The bombardments of uranium and thorium targets were carried out and the numbers of α particles per fission (α/f) were deduced from the measurements as described in the preceding section. The value $(3.35 \pm 0.20) \times 10^{-3}$, which was assumed for the Cf^{252} reference source, was taken from the work of Nobles.⁶ The results are given in Table I and are plotted in Fig. 4 together with Nobles' values of (α/f) for spontaneous and slow neutron fission. Also plotted is the value of Perfilov *et al.*⁷ for 14-MeV neutron-induced fission of uranium. The abscissa in this figure is the excitation

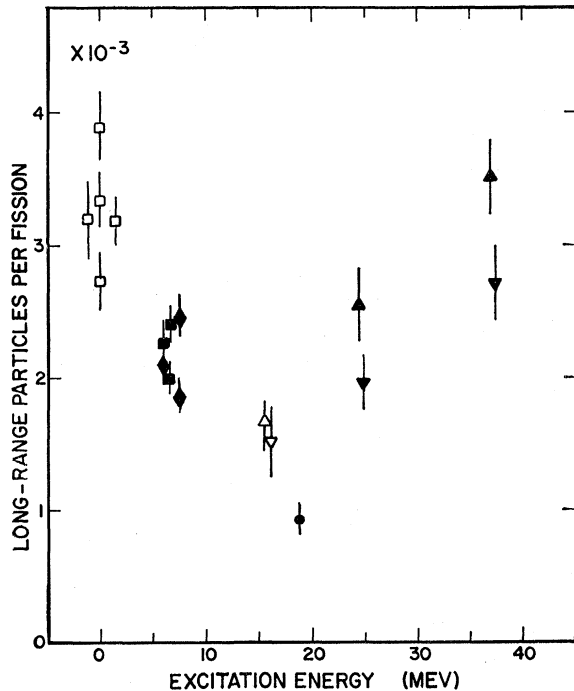


FIG. 4. The number of α particles produced per fission in various bombardments. The squares refer to the work of Nobles. (Ref. 6) Some small liberties were taken with the abscissal location of these data in order to avoid confusion due to overlap. The closed circle is from a 14-MeV neutron bombardment of U^{238} of Perfilov *et al.* (Ref. 7) and the triangles refer to the data of this work. Open triangles are for proton bombardments, closed ones for α -particle bombardments. Inverted triangles refer to Th^{232} bombardments and upright triangles to bombardments of U^{238} .

energy of the original nucleus formed in the bombardment. It is seen from the figure that up to values of about 20 MeV for the initial excitation energy, the value (α/f) falls steeply with energy. It rises again at the higher energies.

Neither the fall nor the rise is understood. This discussion will therefore be limited mainly to qualitative considerations which tend to suggest a direction for further investigations.

Let us begin by considering the points up to 20-MeV excitation. To divide the problem at this energy corresponds to the assumption that the rise at 20 MeV is to be associated with some effect which is still relatively unimportant at the lower energies.

The ratio (α/f) might reasonably be expected to depend on the species of fissioning nucleus, the excitation energy, and perhaps the angular momentum of this nucleus. The angular momentum happens to be zero for all of the examples of spontaneous fission and it is zero or small for all of the other bombardments to the left of 20 MeV in Fig. 4. The problem then is to separate the dependence on the species (say on A and Z) from the dependence on excitation energy. First consider the five points we have for zero energy (spontaneous fission) and imagine that their values, (α/f) , are plotted in three dimensions as a function of Z and A . They are expected to lie on some sort of smooth surface. Because there are only five data points to determine the surface, one must be satisfied with a plane, the plane which is tangent to the actual surface in the middle of the points. (This amounts to being satisfied with the first-order Taylor expansion.) If the surface happens to be very irregular or has great curvatures, it may prove impossible to fit even just five points with a plane. Without more data or a theoretical prescription for the nature of the surface one would in that event probably have to abandon the attempt to fit the data. In the present case, the fitting of the data with a plane turns out to be moderately successful. If the equation for the "best" plane happens to be $(\alpha/f) = a + bZ + cA$, then (α/f) should have a linear dependence on the variable $[(b/c)Z + A]$. The ratio b/c measures the orientation of the gradient of the plane and can be found if one knows or can guess any contour, $(\alpha/f) = \text{constant}$.

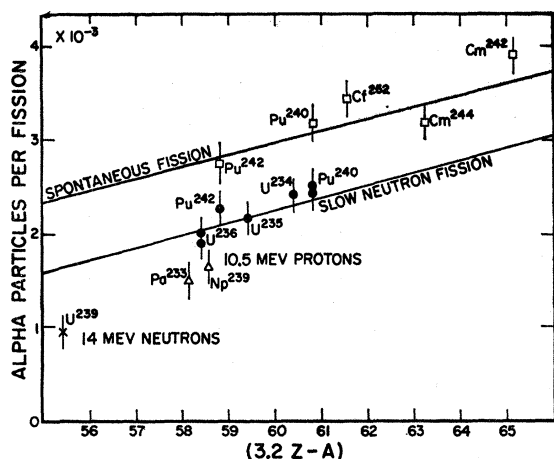


FIG. 5. The number of α particles produced per fission at different excitation energies plotted as a function of the variable $(3.2 Z - A)$ of the compound nucleus formed in the bombardment. The significance of this parameter is discussed in the text.

In this way it was found from an examination of the data points that the "best" value for b/c for the spontaneous fission data was -3.2 and that the same value also happened to fit the data points for slow (thermal or 1 MeV) neutron-induced fission. The plots of (α/f) as a function of $(3.2 Z - A)$ are given in Fig. 5 and it is seen that the data do exhibit a rough linear relationship. One can therefore conclude that (α/f) seems to depend smoothly on the coordinates (Z, A) . The nature of the dependence is such that (α/f) is larger for the more fissionable nuclei. This was pointed out by Nobles,⁶ who plotted his data against the variable Z^2/A , familiar from the liquid drop model. Z^2/A also increases with fissionability. In the region of the periodic table being considered here, the variable Z^2/A , when expanded, corresponds roughly to $(5Z - A)$. This form happens to be not quite as good as $(3.2 Z - A)$ from an empirical point of view. There is, moreover, no theoretical reason that Z^2/A should have anything to do with α -particle emission in fission. It should be safe to use the lines of Fig. 5 for interpolations and short extrapolations to obtain (α/f) values for unmeasured species.

It is useful to use the lines in Fig. 5 to deduce the excitation energy dependence of (α/f) at least for the two species actually studied in the proton bombardments. Consider $U^{238}(p, f)Np^{239}$, for example. The observed fission events consist of fissions of Np^{239} at roughly 16 MeV (the initial excitation energy) plus fissions of Np^{238} at about 7 MeV (i.e., after a neutron is emitted).

The observed value of (α/f) is 1.66×10^{-3} . The value of (α/f) for the Np^{238} fissions can be obtained from the slow neutron curve. It is 2.2×10^{-3} . According to reasonable estimates of fission probabilities,^{15,17} it turns

¹⁷ J. R. Huizenga and R. Vandenbosch, in *Nuclear Reactions*, edited by P. M. Endt and P. B. Smith (North-Holland Publishing Company, Amsterdam, 1962), Vol. II.

out that $\frac{3}{4}$ as many fissions take place from Np^{238} as from Np^{239} . Hence,

$$1.66 \times 10^{-3} = \frac{(1)x + (\frac{3}{4})(2.2 \times 10^{-3})}{1 + \frac{3}{4}},$$

where x is the value of (α/f) in Np^{239} at 16 MeV. The solution is $x = 1.25 \times 10^{-3}$. This value is plotted (Fig. 6) together with the values of (α/f) for Np^{239} for zero and "slow neutron" fission energies which one can get from Fig. 5. It is seen that the number of α particles per fission falls rapidly with increasing excitation energy. A very similar relation would be obtained for Pa^{233} . It is to be emphasized that Fig. 6 purports to give the excitation energy dependence of (α/f) for a single species. Figure 4, on the other hand, merely gives the measured value of (α/f) for a number of nuclei (and their descendants after neutron emission) as a function of the original excitation energy.

It also is possible to try to deduce from Fig. 5 the ratio (α/f) for U^{239} at the original excitation energy of 18.5 MeV.⁷ One must again use information about relative probabilities for fission and neutron emission for the species involved,^{15,17} and one must rely on extrapolations to the left of the data points in Fig. 5. The result is that (α/f) in U^{239} at 18.5 MeV is about 0.4×10^{-3} , but the uncertainty in this estimate is considerable. It can be stated, however, that the three species examined consistently indicate that (α/f) falls fairly rapidly as the excitation energy increases.

It should be stressed perhaps that this conclusion is in part a consequence of the assumptions made in the analysis, namely that (α/f) depends on Z , A , and excitation energy. One might, for example, have assumed instead that only first fissions produce α particles, that fissions following neutron emission do not. In that case we would understand the character of Fig. 4 in terms of the increasing number of nonalpha producing fissions that must occur for higher bombarding energies. We reject this particular hypothesis because it seems to be a poor one. Since fission has always shown itself to be a genuine compound nuclear process, it is hard to see how a "first" fission can be physically distinguishable from "second" fission in the way suggested. This alternative was mentioned to make it clear that it was not *proved* that (α/f) falls as the excitation energy in-

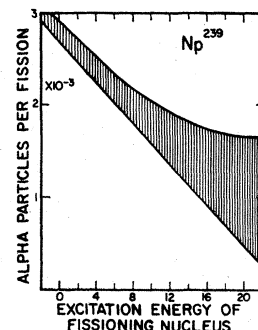


FIG. 6. The number of α particles per fission as a function of the excitation energy of the fissioning nucleus. The width of the curve is supposed to give a rough indication of its uncertainty. The curve was constructed on the basis of the measured rates (α/f) for slow neutron and spontaneous fission of a number of nearby nuclides and the rate in $U^{238}(p, f)Np^{239}$.

creases. This conclusion does, however, follow if one makes what seem to be reasonable assumptions.

One might hope to check this conclusion by careful mass distribution measurements of the coincident fragments since the mass distribution changes sharply with excitation energy. However, such measurements can hardly be expected to provide adequate statistics with present techniques.

If one accepts the conclusion that, at least up to 20 MeV, (α/f) falls with increasing excitation energy, how is one to explain it? Such a question must be answered in terms of a theory for the α -production process in fission. A rough beginning of a theory has been developed that shows some promise for explaining several aspects of the α -production phenomenon in fission other than those being considered now.¹⁸ Briefly in this theory, the α particles appear in the (high Coulomb potential) region between fragments when they manage to acquire sufficient energy from the sudden change in nuclear potential during scission to be "left behind." One of the ingredients in a calculation with this theory is the probability of finding α particles in the neck region of the fissioning nucleus just before the neck snaps. If this probability should decrease as the excitation energy increases, we could perhaps understand the observed (α/f) energy dependence. In some views, clustering of nucleons into α particles is essentially a low excitation energy phenomenon. When the nuclear temperature approaches the cluster dissociation energy, the clusters tend to dissolve. Unfortunately, no detailed calculations are available concerning the energy dependence of cluster formation. The fact that (α/f) rises again (right side of Fig. 4) beyond 20 MeV is not necessarily in conflict with the present suggestion since the cluster dissociation must be gradual (the temperature varies only slowly with energy). If there is some other feature of the α -production phenomenon which strongly favors the production above 20 MeV, there may still be enough clusters around to allow for the observed upturn of (α/f) .

Another feature of the theory upon which the α -particle production rate depends strongly is the intensity of the snap with which the pieces of torn nuclear neck collapse back onto the fragments.¹⁹ If the snap is too slow, the α particles can adiabatically adjust to the changing potential, never acquiring enough energy to leave the rest of the nuclear matter. Two possible reasons for a weakening of the scission snap with increasing excitation energy suggest themselves.

1. One would expect the nuclear viscosity to increase with increasing temperature. If the viscosity becomes large enough, it could lengthen the period of the oscillation associated with the snap. It must, how-

ever, be pointed out that one needs considerable damping (approaching critical damping) before there are appreciable effects on the period. The frequency of an oscillation with damping ω_D is related to the undamped natural frequency ω_0 through

$$\omega_D = \omega_0 [1 - (\Gamma/2\omega_0)^2]^{1/2},$$

where Γ is the width of the resonance curve for the oscillator with damping. In the giant photonuclear resonance, for example, $\Gamma/2\omega_0$ is about one-tenth. In that case there is therefore hardly any slowing down due to damping. One might imagine that the viscous forces in the scission snap are of the same order as those in the photonuclear effect. One would then expect no significant effect on the rate of α -particle production due to viscosity.

2. Another way in which increasing excitation could reduce the snap intensity is through its effect on the tensile strength. If the tensile strength of the nuclear matter in the thin neck between fragments falls off fast enough with increasing excitation energy, there may be a severe reduction at higher energies of the number of long-necked configurations at scission. There is evidence that α particles are produced in those fissions where the distortion at scission is largest, i.e., in just those configurations that would be most affected by a reduction in tensile strength.

There are thus a number of possible reasons for a decrease in α -particle production rate with increasing temperature. Unfortunately, it is difficult to make reliable estimates of the magnitude of these different effects or even to decide their relative importance.

It now remains to consider the upturn in (α/f) beyond 20 MeV. The points showing this upturn were all obtained in α -particle bombardments. On quite general grounds one would, therefore, suggest looking for the effect with other projectiles, say 30-MeV protons. If there were a difference between the results in proton and α -particle bombardments, it could perhaps be connected with the large amount of angular momentum brought into the nucleus by the α particles. If angular momentum effects have a role in determining (α/f) , this could also be established in experiments with α particles alone. The measurements reported here were performed with the fission counter at 135°. If one could look at angles further back towards 180°, one could be looking at fissions from nuclei with higher than average angular momentum J ; towards 90° one would be looking at fissions from nuclei with lower than average J . One could explore a possible J dependence of (α/f) by repeating the present measurement at other angles. One could keep the α counter always at right angles to the fission counter, as is desired in these experiments, by having it looking at the target (properly tilted) directly from above. Such experiments are now under way at the University of Washington.

Another factor possibly involved in the upturn of (α/f) at 20 MeV is the fission mass distribution. It is

¹⁸ I. Halpern, Progress Report, Cyclotron Research, University of Washington, 1961 (unpublished); also, I. Halpern (to be published).

¹⁹ R. W. Fuller, Phys. Rev. 126, 684 (1962).

roughly at this excitation energy that symmetric fission begins to dominate over asymmetric fission.¹⁵ If α emission happens to be more probable in symmetric divisions than in asymmetric ones, one could hope to account for the observations at the higher energies. It should be remarked that the many studies of α -particle emission in fission which have so far been made do not provide reliable information concerning α -particle emission in symmetric divisions because symmetric division is an extremely rare occurrence in fission at the low excitation energies generally studied. Perfilov's study⁷ and this one are the first ones dealing with energies significantly higher than those provided by thermal neutrons. There is a theoretical reason that makes one consider seriously the suggestion that the α -particle emission rate may be larger in symmetric than in asymmetric fission. It would appear that one reason that α -particle production in fission is so rare is that it requires a greater amount of energy to release a fission alpha particle than is ordinarily available at the time of scission.¹⁸ The observation that fragments in symmetric fission have smaller average kinetic energies and larger excitation energies than those in asymmetric fission²⁰ can reasonably be taken to mean that scission in symmetric fission occurs from more stretched configurations. Although the distortion energy at scission is generally converted into internal excitation of the fragments, one must suppose that from time to time it becomes available for alpha-particle release. The amount of this distortion energy appears to be not nearly so marginal for alpha-particle production in symmetric fissions as in asymmetric fissions.

The speculation that more α particles are emitted in symmetric than in asymmetric fission could be tested, in principle, by a careful study of the coincident fission fragment mass distribution or the kinetic-energy distribution. One would, for example, expect to see a difference between the fragment energy spectra observed with and without the requirement of an α -particle coincidence. No such difference is apparent in the present

²⁰ See, for example, H. C. Britt, H. E. Wegner, and J. Gursky, *Phys. Rev. Letters* **8**, 98 (1962).

data [Fig. 3(b)], but the expected difference is not great, and the statistics are poor. Perhaps a simpler way to check whether symmetric fissions are accompanied by more α particles than asymmetric fission is to study α -particle production in fission occurring in bombardments of lead or bismuth. Here the mass distributions happen to be symmetric. One would expect that the ratios (α/f) might be larger than those observed so far and that they would not increase with bombarding energy (Fig. 4) but would instead stay constant or decrease. In view of the small fission cross sections of lead and bismuth, it is at present difficult to measure the α -particle production rates in these fissions.

To summarize the present results: The observed rates of α -particle production in coincidence with fission at bombarding energies below 20 MeV can most simply be interpreted in terms of a decrease in the production rate with increasing excitation energy. It also is found that the production rate is largest for the more fissionable nuclei. The number of coincident α particles per fission increases sharply at (α -particle) bombarding energies above 20 MeV. Possible reasons for the initial decrease and the later increase with energy have been discussed, but none of them receives strong support from presently available observations. Such support possibly could come from a repetition of measurements like those reported here with better statistical accuracy. The accumulation of good statistics is, however, a difficult matter because of the inherently low rates of fission-alpha coincidences. A few qualitative experiments also have been suggested which would tend to explore some possible implications of the work described here.

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