Emission of Alpha Particles in the Fission of U^{238} by 11- to 21-MeV Protons*

T. D. THOMAS† AND S. L. WHETSTONE, JR.
Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico
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The rate of emission of alpha particles in coincidence with fission has been measured for U^{238} bombarded with protons of energies 11, 12, 13, 14, 15, 17, and 21 MeV. The results of these measurements may be summarized by the formula $Y_\alpha = (1.53\pm0.18)+(0.044\pm0.010)E_p$, where Y_α is the number of alpha particles per thousand fissions and E_p is the incident proton energy in MeV. For spontaneous fission of Cf^{252}, Y_α is now known to include $(2\pm1)\%$ of He^6 and possibly much smaller amounts of slightly heavier particles. Y_α increases by about 2.2% per MeV from 11 to 21 MeV, with no evidence for a minimum in this range. The angular correlations between alpha particles and fission fragments have been measured at 11-, 15-, 17-, and 21-MeV proton energy and for the spontaneous fission of Cf^{252}. When corrected for the angular resolution of the detectors, the distributions are found to be approximately Gaussian with means and standard deviations $94\pm0.3°$ (center-of-mass) and $14.7\pm0.4°$ (nearly independent of incident proton energy) for the induced fission, and $93\pm0.6°$ and $12.0\pm0.4°$ for Cf^{252}. Absolute values of Y_α were obtained relative to the absolute rate of emission of alpha particles from the spontaneous fission of Cf^{252} as given in the literature and by an independent measurement reported in this paper.

I. INTRODUCTION

LIGHT charged particles are emitted in binary fission with a probability of from 0.1 to 0.5%.[1] These particles are usually alpha particles but evidence has also been reported for the emission of protons, tritons, He^6, and probably other particles.[2–8] Only the relatively abundant alpha particles have actually been measured in coincidence with fission fragments; all that is known about the rarer particles is their emission probability and the upper portion of their energy spectra. Many studies of the process have not distingushed among these various particles.

Coincidence studies of the alpha particles indicate that they originate in the region between the fission fragments at, or very shortly after, the time of scission.[9,10] Their production is believed to take place in the neck between the separating fragments at a time when the nuclear matter at that location is being "stretched to its limit."[10] The probability for emission of these particles, their kinetic energies, and their direction of emission should depend on the fission fragment configurations at and immediately after scission. Measurements of these quantities and of various correlations between alpha-particle energies, directions of particle emission, and fission-fragment energies and masses should therefore provide much needed information on the scission mechanism.

These observables and correlations can be further studied for a variety of fissioning nuclei, produced by different combinations of target nucleus, projectile, and bombarding energy. At the lower bombarding energies the fissioning nuclei will, in general, be characterized by well-defined nuclear charge, mass, excitation energy, and angular momentum. At higher bombarding energies prefission neutron emission and fission following a direct interaction affect the uniqueness of the properties of the fissioning nucleus. In addition there is a possibility that a second "mode" of fission, characterized by a preference for symmetric fission, may become important.

Since the data on light-charged-particle emission have been collected over a period of more than 20 years of improving techniques, it is not surprising that many of them are contradictory. As an example, we note that reported values of the probability for emission of light charged particles in the thermal-neutron-induced fission of U^{235} vary from 0.002 to 0.004 per binary fission. We have compiled in Appendix II the available data on this probability as a function of excitation energy. In Fig. 1 we have plotted selected data from this compilation to illustrate the general features of the data. Included are all of the results on the initial compound nucleus U^{236}, which has been studied over the excitation range from 6.5 to 37 MeV, and part of the data for Pu^{240}, to illustrate what is known for excitation energies between 0 and 6.5 MeV.

The decrease in probability from zero excitation energy to 6.5 MeV is apparently real. The yield of light charged particles from both the spontaneous fission of Pu^{240} and the thermal-neutron-induced fission of Pu^{239} has been measured by Mostovaya[11] and by Nobles.[12]

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† On leave from Department of Chemistry, Princeton University, Princeton, New Jersey.
1 For a recent review of this subject see E. K. Hyde, *The Nuclear Properties of the Heavy Elements* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1964), Vol. III, pp. 131–140.
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10 I. Halpern, in *Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965), Vol. II, p. 369.
11 T. A. Mostovaya, reported by V. I. Mostovoi, in *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy* (United Nations, New York, 1956), Vol. 2, p. 226.
12 Ralph A. Nobles, Phys. Rev. **126**, 1508 (1962).

Both found that the nuclei with higher excitation energy give fewer light charged particles. This drop in yield, together with the very low value reported by Perfilov, Solov'eva, and Filov[13] for U^{236} at an excitation energy of 20.4 MeV, has led to the conclusion that the probability for light-charged-particle emission falls rapidly with increasing excitation energy of the fissioning nucleus. Results from alpha-particle-induced fission by Coleman, Fairhall, and Halpern[14] indicate that this probability does not continue to decrease, but, at an excitation energy of about 20 MeV, begins to rise, and at 37 MeV reaches a value even higher than the value for thermal-neutron-induced fission.

Recent measurements by Drapchinskii, Kovalenko, Petrzhak, and Tyutyugin[15] for U^{235} plus 14-MeV neutrons indicate that the result obtained by Perfilov *et al.* may be in error and that the probability for light-charged-particle emission is nearly the same at an excitation energy of 20 MeV as at 6.5 MeV. Furthermore, Coleman *et al.* did not make any correction for the possibility of the $(\alpha,\alpha' f)$ reaction, which they could not distinguish from the $(\alpha,f\alpha)$ reaction. In a private communication, Fairhall[16] has informed us that the values reported in their paper[14] will be considerably lowered if a correction is made for this effect.

We may summarize the above by saying that there has been considerable uncertainty as to the excitation-energy dependence of light-charged-particle emission in fission. With a view toward clarifying this situation we have measured the probability for alpha-particle emission in fission for the initial compound system Np^{239} produced by bombarding U^{238} with protons of energies from 11 to 21 MeV, corresponding to excitation energies in Np^{239} from 16.2 to 26.2 MeV.

The choice of projectile was based on the expectation that the $(p,\alpha f)$ reaction, in which an alpha particle is emitted before fission, would be relatively less important than the $(\alpha,\alpha' f)$ reaction. Measurements were made down to a bombarding energy of 11 MeV, where, because of the Coulomb barrier, the probability of forming the compound nucleus was small.

Since our experiments were made with limited solid angles for the detection of the alpha particles and fission fragments and with a low-energy cutoff on the spectrum of alpha particles detected, it was necessary to measure the angular correlation between the alpha particles and fission fragments and the energy spectrum for the alpha particles to see if these changed appreciably with bombarding energy. The angular correlation measurements also made it possible to estimate the relative importance of the otherwise indistinguishable contribution to the coincident alpha-particle production from the $(p,\alpha f)$ reaction.

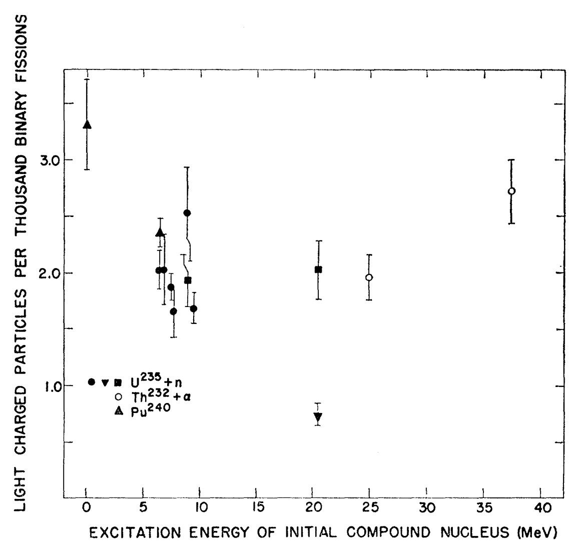

FIG. 1. Representative data on the emission of light charged particles in fission as a function of excitation energy of the initial compound nucleus prior to our results (cf. Fig. 6). Open circles are from Ref. 14; solid squares, Ref. 15; inverted triangle, Ref. 13. For others, see Table VI, Appendix II.

A straightforward way to obtain absolute values for the alpha-to-fission ratio for the total solid angle and energy spectrum is to make measurements with Cf^{252}, for which this ratio is known (see Appendix I), using the same equipment. Since other work[17] has indicated a difference between the angular correlations for Cf^{252} and proton-induced fission of U^{238}, it was particularly important in this case to make careful measurements of this correlation with our equipment for Cf^{252} and for proton-induced fission at least at one bombarding energy.

II. EXPERIMENTAL PROCEDURES

Our experiment was designed to measure as a function of incident proton energy the coincidence rate between single fission fragments and energetic alpha particles in a fixed counting geometry. The alpha particles were detected in a ΔE-E counter telescope fixed at 135° to the beam for all of the measurements. Two detectors, fixed approximately 20° apart, were used to detect fission fragments. The use of two fission detectors permitted more rapid and possibly more accurate measurements of the alpha-particle, fission-fragment angular correlations. To accumulate data on the energy dependence of the alpha-to-fission ratio, one of the detectors was fixed at about 90° to the alpha detector to take advantage of the relatively large counting rate there; the other was fixed at about 70°. These angles correspond to 135° and 155° to the beam.

[13] N. A. Perfilov, Z. I. Solov'eva, and R. S. Filov, At. Energ. (USSR) **14**, 575 (1963).

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

[15] L. V. Drapchinskii, S. S. Kovalenko, K. A. Petrzhak, and I. I. Tyutyugin, At. Energ. (USSR) **16**, 144 (1964).

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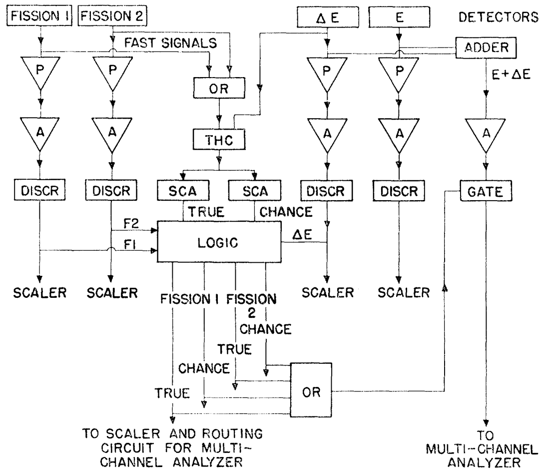

FIG. 2. Schematic drawing of electronics. The symbols have the following significance: P—preamplifier, A—linear amplifier, THC—time-to-pulse-height converter, DISCR—discriminator, SCA—single-channel analyzer.

Beam

Protons were accelerated to energies from 11 to 17 MeV in the Los Alamos Model FN Tandem Van de Graaff and to energies from 15 to 21 MeV in this machine with beam injected from the Los Alamos Vertical Van de Graaff. The beam was momentum-analyzed in a 90° spectrometer magnet, focused by a quadrupole magnet, and passed through collimating slits $\frac{1}{8}$ in. high by $\frac{1}{16}$in. wide into a 20-in.-diam reaction chamber. The target was replaced occasionally with a piece of photographic paper to verify that the beam passed through the axis of rotation of the chamber and maintained proper focus. The beam spot at the target position was of approximately the same dimensions as the collimating aperture. Beam currents in the chamber ranged from 50 to 300 nA.

Target and Source

The target was 500 μg/cm^2 of uranium (U^{238}) oxide deposited by evaporation from a hot filament onto a thin carbon film. Protons, fission fragments, and long-range alpha particles pass through the target and backing with energy loss insignificant with respect to the requirements of the experiment. The Cf^{252} source, about $\frac{1}{8}$ in. in diameter, mounted on 0.003-in. platinum foil and covered with a 100-μg/cm^2 nickel foil, provided about 10^5 fissions/min for calibration.

Detectors

The fission detectors were *p-n* junction semiconductor devices, approximately 1 cm sq and self-collimated, with each subtending an angle of about 14° at the center of the chamber. They were operated with the front surface grounded and with negative bias applied to the back surface. (If positive bias was applied to the front surface, a high current of electrons from the target was attracted to this surface during irradiation.) By the time these detectors had been struck by 10^8 fission fragments, the leakage current at 50-V bias had risen from 1 to 15 μA. At this time they were replaced with a new pair.

The light charged particles were detected in a ΔE-E counter telescope consisting of two semiconductor detectors: a totally depleted "ΔE" detector, 112 μ thick, followed by an "E" detector, depleted to depth of about 400 μ. An aluminum foil with a surface density of 6.6 mg/cm^2 prevented fission fragments and low-energy electrons from reaching the detectors, yet transmitted alpha particles with energies greater than 5.5 MeV. A 0.75-cm-sq collimator was placed in front of this system to ensure that all charged particles passing through the ΔE detector would, if unscattered, strike the E detector and that no charged particles could reach the E detector without passing through the ΔE detector. The angle subtended at the center of the chamber was about 14°. The maximum energy deposited by protons, deuterons, and tritons in the ΔE detector was 3.2, 4.3, and 4.8 MeV, respectively. By accepting only particles that deposit more energy than 4.8 MeV in the ΔE detector, we were able to discriminate against all of these lighter particles (except when two of the abundant protons passed through the ΔE detector at about the same time). Alpha particles leaving the aluminum absorber with energies between 4.8 and 30 MeV deposited more than 4.8 MeV in the ΔE detectors. Approximately 90% of the alpha particles of interest[12] and essentially none of the singly charged particles met these requirements.

The detectors were mounted on arms with angular positions that could be precisely controlled from outside the chamber. The calibration of these controls was determined by rotating each detector into the line of sight of a transit that had been previously aligned to the collimator aperture and chamber center. The calibration was reproducible to about ±0.5 deg.

Electronics

The electronics for the experiment included a time-to-height converter system to determine that a fast coincidence had occurred between the charged-particle detector and one of the fission detectors, a charged-particle identification system to determine whether an acceptable charged particle had been detected, a logic system to decide (1) which of the two fission detectors had detected a fragment, and (2) whether the coincident events were within about 10 nsec of each other (true coincidence) or were separated by some greater time (chance coincidence). There were four possible outputs from the logic circuit corresponding to either true or chance events in either of the two fission detectors. One multichannel analyzer recorded the output of the time-to-height converter in order to show whether there was any drift in the timing. Another analyzer recorded in each of four quadrants the spectrum of particles from the charged-particle detector according to which of the four logical conditions was satisfied. A schematic drawing of the apparatus is shown in Fig. 2.

Fast signals were developed by passing the charge collected from the detectors through ORTEC time-pickoff units. A Chronetics Fan-in circuit was used to provided a carefully shaped pulse to one input of the time-to-height converter if either of the fission detectors detected a fragment and another pulse of identical shape to the other input when a particle was detected by the ΔE detector. The time resolution of this system was less than 10 nsec for true coincidences. True-coincidence events produced outputs of nearly maximum pulse height from the converter; all other pulse heights corresponded to chance events. Two single-channel analyzers at the output of the converter were used to determine whether an event was a true or chance coincidence. In order to improve statistics we made the chance channel six times as wide as the true channel.

Signals from the various semiconductor detectors after passing through the time-pickoff units were converted to voltage pulses in charge-sensitive preamplifiers. These pulses were amplified by double-delay-line-clipped linear amplifiers. The signals from the E and ΔE preamplifiers were summed and amplified to give a signal proportional to the total energy of the charged particle.

Four coincidence circuits were used to determine whether a particular event was a true or chance coincidence, whether the ΔE detector had produced a large enough pulse, and which fission detector had been struck by a fragment. The outputs of these four coincidence units were sent to four scalers, to the routing circuit of one of the multichannel analyzers, and to a fourfold "Or" circuit. The output of the "Or" circuit opened a linear gate, allowing the $E+\Delta E$ pulse to go to the input of the multichannel analyzer, the routing signal directing the pulse to the proper quadrant of the analyzer. The information stored was, thus, the energy spectra of charged particles in true and chance coincidence with each fission detector, the total number of true and chance events between the charged-particle detector and each fission detector, and the total number of fission events in each fission detector. In addition, the singles rates in the ΔE and E detectors, the number of gates, and the integrated beam current were monitored.

The Measurements

Measurements were made at proton energies of 11, 12, 13, 14, 15, 17, and 21 MeV during three separate running periods. Most of the apparatus had to be set up anew for each period. During the third period, the fission detectors and E detector were replaced. During each period, measurements were made at 15 MeV in order to check the reproducibility of the experiment from one period to the next. After the first two periods, calibration runs were made with Cf^{252}, using the same equipment.

TABLE I. Angular-correlation results.

Proton energy (MeV)	$\theta_{\alpha f}$ (deg)	Relative coincident rate
	First running period	
11	70	0.280±0.079
	89	1.
	110	0.778±0.082
	129	0.082±0.041
14	70	0.263±0.029
	89	1.
15	41	0.040±0.015
	60	0.176±0.022
	70	0.296±0.023
	89	1.
	110	0.735±0.042
	129	0.099±0.015
	160	0.045±0.012
	179	0.079±0.014
	Second running period	
11	71	0.397±0.056
	90	1.
12	71	0.419±0.058
	90	1.
13	71	0.404±0.041
	90	1.
15	81	0.815±0.057
	90	1.
	100	0.895±0.061
	109	0.613±0.041
	150	0.023±0.013
	169	0.008±0.013
	Third running period	
15	70	0.35 ±0.03
	71	0.34 ±0.03
	90	1.
17	70	0.29 ±0.03
	71	0.34 ±0.02
	90	1.
	111	0.76 ±0.04
	130	0.099±0.018
21	70	0.28 ±0.02
	90	1.
	110	0.68 ±0.04
	130	0.087±0.017
	150	0.052±0.011
	170	0.079±0.012
	Cf^{252}	
	71	0.278±0.022
	90	1.
	100	0.959±0.068
	119	0.151±0.024

The beam intensity was controlled so that the chance rates were small compared to the true rates. In some cases, where the true rate itself was small, the true-to-chance ratio was allowed to approach unity; in a few extreme cases this ratio was near zero.

At each energy, measurements were made with fission detectors at about 90° and 70° with respect to the charged-particle detector. At 11, 15, 17, and 21 MeV and for the spontaneous fission of Cf^{252}, measurements

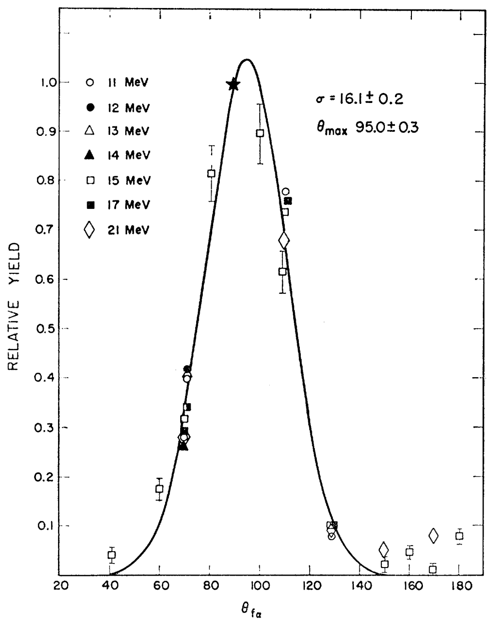

FIG. 3. Angular correlation between alpha particles and fission fragments for different proton energies (See Table I.) All data have been normalized to unity at 90°. The solid curve, fit to the data by least squares, is Gaussian with the indicated parameters. No correction has been made to these data for detector resolution. Gaussian parameters for the intrinsic correlation function are given in the text.

at additional angles were made. The purpose of the measurements at angles other than 90° was fourfold: first, to determine whether the angular correlation for U^{238} plus protons depended on bombarding energy; second, to compare this angular correlation with that for Cf^{252}; third, to investigate the contribution of the $(p,\alpha f)$ reaction to the coincidence rate by making measurements at angles where ternary fission is presumably rare; and fourth, to investigate further the secondary maximum in the coincidence rate reported by Atneosen, Thomas, and Garvey[17] for an angle of 180° between the fission-fragment detector and the alpha-particle detector.

III. RESULTS

Dependence of the Alpha-Particle Emission Rate on Bombarding Energy

We consider the possible dependence on the bombarding energy of the angular correlation between fission fragments and alpha particles.

In Fig. 3 we have plotted the results, also given in Table I, of the angular-correlation measurements. All are normalized to unity at 90°. The curve shown in Fig. 3 is Gaussian, fitted to the data by least squares, each datum point weighted according to the inverse square of its standard deviation. To exclude the small peak evident near 180°, data for $\theta_{f\alpha}\geq 150°$ were not included. The distribution peaks at 95°; correction for center-of-mass motion shifts the position of the maximum to about 94°. This value is in good agreement with that found by Atneosen *et al.*,[17] but is still about 1.5° greater than the value calculated from conservation of momentum. The width of the distribution is, however, considerably narrower than that reported by Atneosen *et al.*, whose measurements were, however, handicapped by poor counting statistics.

TABLE II. Gaussian parameters derived from angular-correlation measurements.

Energy	$\theta_{\max}$	σ
11	95.2±1.0	16.3±0.8
15	94.2±0.5	16.2±0.3
17	96.1±0.5	16.3±0.4
21	95.3±0.02	15.5±0.01
All data	95.0±0.3	16.1±0.2

To investigate more closely possible energy dependence of the angular correlation, we have also fit Gaussian curves to the data for 11, 15, 17, and 21 MeV, individually. The results are summarized in Table II. The errors quoted are calculated from the deviations of the measured points from the calculated line. There is a 50% probability that the true value of the parameters lies more than one deviation away. The tabulated quantity σ is the usual width parameter of the Gaussian distribution. Although the individual $\theta_{\max}$ and σ scatter about their average values, there does not seem to be evidence for a significant variation in the angular correlation as the energy is varied.

We next consider the fraction of alpha particles too low in energy (<8.8 MeV) to trigger the lower level discriminator in the charged-particle detection system after passage through the absorber foil. From the energy spectra of the alpha particles detected we have calculated the average particle energy and the root-mean-square deviation from the average for incident proton energies of 15, 17, and 21 MeV. The results are given in Table III. Since only an approximate energy calibration was made, no significance should be attached to the absolute values in this table. The values listed differ from one another by somewhat more than we would expect from the experimental uncertainties, indicating a possible variation in the spectrum of long-range alpha particles with incident proton energy. To evaluate the effect of such a variation on the experimentally determined probability for alpha emission in fission, we

TABLE III. Average energies and rms deviations from the average for coincident alpha particles at different bombarding energies. (Absolute energy scale is only approximate.)

Bombarding energy (MeV)	Average alpha-particle energy (MeV)	rms deviation from the average
15	15.37±0.18	5.80
17	15.06±0.17	5.80
21	14.68±0.16	5.99
Average	15.00±0.10	5.88

make the following calculation: We assume for this purpose that the spectrum can be crudely described by a Gaussian function, peaked at the average value, with the value of the standard deviation taken from the average given in Table III and normalized to unity. We calculate that for the average of the three measurements, 86% of the alpha particles have energies greater than 8.8 MeV. (From Nobles's data[12] on Cf^{252} we estimate this quantity to be 91%.) For the three energies, 15, 17, and 21 MeV, we calculate 87%, 86%, and 84%, respectively. The root-mean-square deviation of these values from their average is 1.4%, which we take as the error in our measurements due to changes in the spectral shape as a function of incident proton energy. This error is small compared to others, and we ignored it.

As an aside it should be mentioned that although our energy spectra for $\theta_{\alpha f}=90°$ and for $\theta_{\alpha f}=110°$ were similar in shape to that observed for Cf^{252}, the energy spectrum for $\theta_{\alpha f}=70°$ appeared with high statistical significance to be composed of two broad peaks, separated by about 6 MeV. The high-energy edge of this spectrum appeared at an energy higher than the other spectra by about 3 MeV. This is not easily explanable from the $\theta_{\alpha f}$, E_α correlation measured for Cf^{252} (Ref. 18) but may reflect a possible dependence of the spectrum on the angle with respect to the light (or heavy) fragment.

Our measurement of the alpha-particle, fission-fragment coincident rate with both detectors at fixed angles with respect to the beam may not represent an unbiased sample of the coincidence rate averaged over all angles to the beam because of a possible dependence of the probability for alpha-particle emission on the fission-fragment direction. Such a dependence might result from the fact that fragments emitted at different angles to the beam arise from different distributions of angular momentum in the compound nucleus. Since the angular momentum distribution depends on the energy of the incident particle, such a dependence could lead not only to an erroneous absolute value of the probability for alpha-particle emission in fission (averaged over all angles) but also to an apparent dependence of this probability on the energy of the bombarding particle.

The angular distribution of fission fragments accompanied by light charged particles has been measured by Atneosen *et al.*[17] for 17.5-MeV protons incident on U^{238} over the range 90° to 150° with the two detectors 90° apart. Within the statistical uncertainties of ±15%, their results indicate that the probability for light-charged-particle emission in fission does not depend on the angle between the fission-fragment direction and the beam, over the angular range studied. The result of Ramanna, Nair, and Kapoor[19] for 14-MeV neutron-induced fission of U^{238}, which indicates an extremely strong preference for alpha-particle emission at 0° and 180° with respect to the beam, is difficult to accept. A more recent result of Hattangadi, Methasiri, Nadkarni, Ramanna, and Rama Rao[20] for 3-MeV neutron induced fission of U^{235} gives a much smaller anisotropy of alpha-particle emission:

$$N(0°)/N(90°)=1.32\pm0.12.$$

The angular distribution of binary fission fragments for 3-MeV neutron-induced fission of U^{235} (Ref. 21) can be approximated by

$$W(\theta)=(1+0.17\cos^2\theta)/2.113(2\pi),$$

which is normalized to unity when integrated over the sphere. The results obtained by Hattangadi *et al.*[20] are consistent with an angular distribution for fission fragments accompanied by light charged particles given by

$$W(\theta)=Y(1-0.375\cos^2\theta)/1.75(2\pi),$$

which is normalized to Y, the total probability for light-charged-particle emission in fission, when integrated over the sphere. The ratio of these two expressions evaluated at $\theta=135°$, the angle of our measurements, is 0.90 Y, 10% lower than the ratio of the total probabilities. Unless there is an extremely rapid variation in the angular-distribution parameters with proton bombarding energy, this error will depend only weakly on the incident proton energy.

As was pointed out by Coleman *et al.*,[14] the detection of fragments at 135° to the beam will provide a sample of those nuclei with spins nearly equal to the average spin of all the compound nuclei formed.

Since we observed no appreciable change in the alpha-particle energy spectra or in the alpha-fragment angular

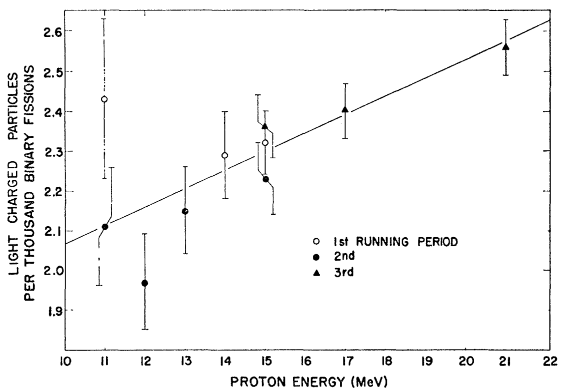

FIG. 4. Dependence of the number of light charged particles per thousand fissions on the bombarding-proton energy. Errors shown are relative and do not include the error involved in determining the absolute probability. The straight line was obtained by a least-squares fit to the data listed under "Coincidences per 10^7 fissions, weighted average" of Table IV.

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TABLE IV. Summary of measurements at $\theta_{\alpha f}=90°$.

E_p, proton energy (MeV)	N_c coincidences per detected fission ($\times 10^7$) First running period	Second	Third	Weighted average	Y_α, alpha-particle emission probability ($\times 10^3$)[b]	Y_{LC}, light-charged-particle emission probability ($\times 10^3$)[c]
11	261±21[a]	227±16		239±13	2.12±0.16	2.25±0.17
12		212±13		212±13	1.88±0.15	1.99±0.16
13		231±12		231±12	2.04±0.15	2.16±0.16
14	246±12[a]			246±12	2.18±0.16	2.31±0.17
15	249± 8[a]	240±10	254±8	249± 5	2.20±0.13	2.33±0.14
17			258±7	258± 7	2.28±0.14	2.42±0.15
21			275±8	275± 8	2.43±0.15	2.58±0.16
Cf^{252}	454±27[a]	441±20		445±16		

[a] These measurements, actually taken at 89°, have been increased by 2%.
[b] Converted to absolute yield relative to Cf^{252} measurements by $Y_\alpha=(G'/G)(N_c/N_c')Y_\alpha'=(88.5\pm4.8)N_c$. See text.
[c] $Y_{LC}=(1.06\pm0.01)Y_\alpha$, to include singly charged particles, principally tritons (Ref. 22).

correlation with incident proton energy, no corrections for these effects were made to the data measured with the detectors 90° apart to obtain the energy dependence of the relative number of coincident alpha particles per fission. These data are given in Table IV and plotted in Fig. 4. The uncertainties indicated are those due to the counting statistics for both "true" and "chance" events and to the uncertainty (±5%) in the relative time intervals over which these were counted. Results for the three separate running periods are identified to give a feeling for the reproducibility of the results. The straight line, derived from a weighted least-squares fit to the data points, is

$$N_c(\times 10^7)=(172.6\pm18.1)+(4.97\pm1.14)E_p,$$

where N_c is the number of alpha particle-fission fragment coincidences per fission fragment detected and E_p, the proton energy in MeV.

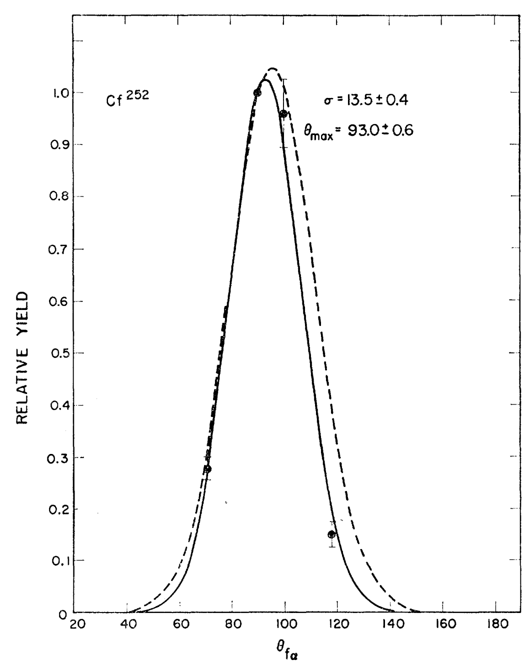

FIG. 5. Angular correlation between alpha particles and fission fragments for Cf^{252}. (See Table I.) Normalized to unity at 90°. The solid curve, fit to the data by least squares, is Gaussian with the indicated parameters. The dashed curve, representing the correlation for U^{238} plus protons, is taken from Fig. 3. No correction has been made to these data for detector resolution. Gaussian parameters for the intrinsic correlation function are given in the text.

Absolute Value of the Probability for Alpha-Particle Emission in Fission

The relative values of the probability for alpha-particle emission in the fission of U^{238} by protons of various energies can be converted to approximate absolute values by a comparison of the coincidences per fission numbers given in Table IV with the number measured under the same instrumental conditions for Cf^{252}. The measured number of alpha-particle–fission-fragment coincidences C, for a given system can be expressed by

$$C=N_fY_\alpha G, \quad (1)$$

where N_f is the number of fission fragments detected, Y_α the probability for alpha-particle emission in fission, and G a geometric factor which takes into account the particular solid angles subtended by the detectors at the source of fissioning nuclei and the angular correlation between fission fragments and alpha particles. Distinguishing by primes the quantities pertinent to Cf^{252} and defining $N_c=C/N_f$ the number of coincidences per fission fragment detected we obtain

$$Y_\alpha=(G'/G)(N_c/N_c')Y_\alpha'. \quad (2)$$

We discuss first the ratio G'/G, which would be unity if the alpha particle-fission fragment angular correlations were the same for the two fissioning systems. In Fig. 5 we show the results of our measurements of the angular correlation for Cf^{252}, which were performed in the same way as were those for U^{238}, but with the uranium target and the proton beam replaced by a Cf^{252} source. The dashed curve is taken from Fig. 3 and represents the data obtained with U^{238} plus protons. The solid curve is a Gaussian fit to the californium data. There is a distinct difference between the angular correlations for the two systems. A similar effect was seen by Atneosen *et al.*[17] although the difference they observed between the two correlations was much greater than it is in ours.

Although we could, in principle, calculate the geometric factor G from the measured angular correlations and the dimensions and positions of the detectors and, hence, use expression (1) to determine Y_α, it is, in practice, difficult to determine the solid angles with sufficient accuracy. We prefer to use expression (2) and calculate the ratio G'/G, which depends only to second order on the solid angle.

To determine the geometric ratio from the measured angular correlations we assume that: (1) The angular correlation between fission fragments and alpha particles is a Gaussian function with its maximum at 94° in the c.m. system (95° in the lab) for Np^{239} and 93° for Cf^{252}. (2) The angular response of each detector is a rectangular function with a width of 14°; the angular response of two detectors folded together is than a triangular function with a base width of 28° (provided that they are separated by an angle $>14°$ and $<166°$). If these assumptions are adequate, we should be able to approximate the measured angular-correlation function by folding the angular-response function into the intrinsic angular-correlation function. We adjust the width parameter of the Gaussian angular-correlation function until the result of the convolution agrees with the Gaussian curves fitted to the measured angular correlation. These convolutions are equivalent to the integration of the intrinsic angular correlation over the solid angle of the detectors and, therefore, yield values of the geometric factor G. This method is essentially the same as that used by Atneosen *et al.*[17]

The data obtained in the bombardment of U^{238} plus protons were best fit with an undispersed angular-correlation function having a standard deviation of 14.7 $\pm 0.4°$ ($35\pm 1°$ full width at half-maximum, FWHM). This is to be compared with the value of $47\pm 12°$ FWHM reported by Atneosen *et al.* We obtained a value of $12.0\pm 0.4°$ ($28\pm 1°$ FWHM) for Cf^{252}, to be compared with their value of $33\pm 2°$ (FWHM). The ratio G'/G of the geometric factor for Cf^{252} to that for U^{238} plus protons determined from these calculations is 1.205 ± 0.031.

The value of Y_α', the probability for alpha-particle emission in the spontaneous fission of Cf^{252}, has been obtained from a critical evaluation of results reported in the literature, including the result of a measurement of our own. We find $Y_\alpha' = (3.27\pm 0.10)\times 10^{-3}$; see Appendix I for details. For spontaneous fission of Cf^{252} it is now known that Y_α' includes $(2\pm 1)\%$ of He^6, and possibly, though in much smaller proportion, other slightly heavier particles.[8]

We obtain for the probability Y_α of alpha emission in the fission of U^{238} by protons under our experimental conditions:

$$Y_\alpha = (1.205\pm 0.031)(3.27\pm 0.10)(\times 10^{-3})(N_c/N_c') = (88.5\pm 4.8)N_c$$

taking from Table IV the value $N_c' = (445\pm 16)\times 10^{-7}$. The values of N_c and the results calculated for Y_α are given in Table IV. The weighted least-squares fit to the induced-fission data gives

$$Y_\alpha = (88.5\pm 4.8)[(172.6\pm 18.1) + (4.97\pm 1.14)E_p](\times 10^{-7}) = [(1.53\pm 0.18) + (0.044\pm 0.010)E_p](\times 10^{-3}),$$

where E_p is the bombarding-proton energy in MeV.

The ratio of the yield of all light charged particles to the yield of alpha particles in the spontaneous fission of Cf^{252} is known to be 1.06 ± 0.01, where the increase is due chiefly to tritons.[22] Assuming the same ratio to apply for induced fission we obtain for the probability Y_{LC} of light-charged-particle emission in the fission of U^{238} by protons:

$Y_{LC} = (1.06\pm 0.01)Y_\alpha$. These results are also given in Table IV. Finally, the weighted least-squares fit gives

$$Y_{LC} = [(1.62\pm 0.19) + (0.047\pm 0.011)E_p](\times 10^{-3}).$$

IV. CONCLUSIONS

The data presented in Fig. 4 indicate that the probability for alpha-particle emission in binary fission increases by about 20% as the proton energy increases from 11 to 21 MeV (corresponding to an excitation-energy range from 16.2 to 26.2 MeV). Part of this increase may be due to contributions from the $(p,\alpha f)$ reaction. At an excitation energy of 16 MeV it is almost impossible for an alpha particle to be emitted with a sufficiently low kinetic energy that the residual nucleus can have enough energy to fission. At 26 MeV, however, such a process is quite possible. A contribution to the coincidence rate from the $(p,\alpha f)$ reaction should be most evident at angles well away from the peak of the alpha-particle, fission-fragment angular correlation. Figure 3 indicates a value of about 3% of the maximum yield in the region around 150 to 160° for the 15-MeV data and perhaps 5% for the 21-MeV data. If the probability for the $(p,\alpha f)$ reaction is more or less independent of the angle between the alpha-particle and fission-fragment direction, then no more than about 5% of the increase in probability from 11 to 21 MeV can be due to this reaction.

Because of the possibility of neutron emission before fission, each of our measurements represents an average probability for light-charged-particle emission taken over a number of fissioning nuclei at different excitation energies. Using the method given by Coleman *et al.*[14] we can extract from our data some approximate infor-

[22] From Cf^{252} there are about 6.5 H^3 per 100 He^4, Refs. 4, 5, 7; for thermal-neutron-induced fission of U^{235}, 4.5 H^3 per 100 He^4, Refs. 2, 3, 6.

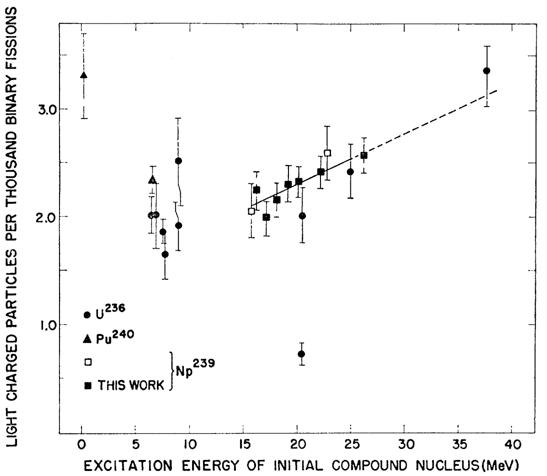

FIG. 6. Dependence of the number of light charged particles per thousand fissions on the excitation energy of the initial compound nucleus. Solid squares are from this work, solid circles from Fig. 1, and open squares from Refs. 14 and 17. The values from Refs. 14 and 17 have been increased by 20% to take into account the different angular correlations for Cf^{252} and the systems studied. The line is from Fig. 4.

mation on the variation of this probability with excitation energy for a single compound nucleus. From Jackson's[23] evaporation model as modified by Vandenbosch, Thomas, Vandenbosch, Glass, and Seaborg[24] together with values of Γ_f/Γ_n from Vandenbosch and Huizenga[25] we can estimate the probabilities of first, second, and third chance fission at each bombarding energy. For 11-MeV protons incident on U^{238}, about two-thirds of the fissioning nuclei are Np^{239} with an excitation energy of 16 MeV and one-third are Np^{238} at about 8 MeV. Coleman *et al.*[14] found from an empirical correlation that the probability for light-charged-particle emission increases by about 0.13×10^{-3} as the quantity $3.2Z\text{-}A$ increases by 1. Thus, for the fissioning nucleus Np^{238} at an excitation energy of 8 MeV we estimate a probability of 2.2×10^{-3} from the data for U^{235} plus thermal neutrons. Taking these numbers, together with our value at a proton energy of 11 MeV of 2.11×10^{-3} for the probability averaged over the two fissioning nuclei, we determine the probability for light-charged-particle emission in the fission of Np^{239} at an excitation energy of 16 MeV to be 2.1×10^{-3}. Using the empirical relationship from the work of Coleman *et al.*[14] we obtain 2.0×10^{-3} for the same nucleus at an excitation energy of 7 MeV. These values are well within experimental error of one another. Similarly, from our data shown in Fig. 4 and the estimates above we estimate an emission probability of 2.7×10^{-3} for Np^{239} at an excitation energy of 24 MeV. Thus at excitation energies 8, 16, and 24 MeV, the probabilities for the emission of light charged particles in the fission of Np^{239} nuclei are 2.0, 2.1, and 2.7×10^{-3}, respectively. If, for example, our measured value (interpolated from our curve of Fig. 4) for the over-all probability for Np^{239} is arbitrarily increased by 5%, the respective probabilities at 8-, 16-, and 24-MeV excitations of fissioning Np^{239} become 2.0, 2.2, and 2.6. It would seem quite likely, therefore, that the probability of light-charged-particle emission in the fission of single nuclear species does increase with excitation energy in the range 16 to 24 MeV, and that there is a minimum in this probability between about 8 and 16 MeV.

Only two other measurements have been reported for the system U^{238} plus protons.[14,17] The compound nucleus U^{236}, however, obtained either from U^{235} plus neutrons or from Th^{232} plus alpha particles, differs from Np^{239} by less than 0.5% in the quantity $3.2Z\text{-}A$. The probability for light-charged-particle emission in fission for these two nuclei should therefore be almost the same.[14] Figure 6 shows the results for the light charged particles per thousand binary fissions as a function of excitation energy of the initial compound nucleus for U^{236}, Np^{239}, and Pu^{240}. We have replotted the data of Fig. 1, but have increased all of the values reported by Coleman *et al.*[14] by 20% to take into account a difference in angular correlation between U^{236} and Cf^{252} of the order of what we have found between Np^{239} and Cf^{252}. In addition, we have plotted as open squares the results obtained by Coleman *et al.*[14] for 10.5-MeV protons incident on U^{238} and by Atneosen *et al.*[17] for 17.5-MeV protons incident on U^{238}. These values have also been corrected for an angular-correlation effect of the magnitude found in our present work. Our data are shown as solid squares. The solid line is taken from the least-squares fit of Fig. 4.

We note that there is fairly good agreement between our results and other data in the same energy region. The probability for light-charged-particle emission appears to go through a minimum at an excitation energy between 9 and 15 MeV. Furthermore, the dashed extension of the line representing our data passes very close to the point determined by Coleman *et al.*[14] for alpha particles incident on Th^{232} corresponding to an excitation energy of 37 MeV in the initial compound nucleus. Their results from U^{238} bombarded by alpha particles lie higher, but must be accepted with caution until more is known about the contributions from the $(\alpha,\alpha' f)$ reaction.

A possible explanation for the rising probability of light-charged-particle emission in fission with increasing excitation energy is found in a suggestion made by Coleman *et al.*[14] that this increase is due to the higher probability for symmetric fission at the higher excitation energies. Britt and Whetstone[26,27] have measured

[23] J. D. Jackson, Can. J. Phys. 34, 767 (1956).
[24] R. Vandenbosch, T. D. Thomas, S. E. Vandenbosch, R. A. Glass, and G. T. Seaborg, Phys. Rev. **111**, 1358 (1958).
[25] R. Vandenbosch and J. R. Huizenga, in *Proceedings of the Second United Nations Conference on the Peaceful Uses of Atomic Energy* (United Nations, Geneva, 1958), p. 284.
[26] H. C. Britt and S. L. Whetstone, Jr., Phys. Rev. 133, B603 (1964).
[27] S. L. Whetstone, Jr., Phys. Rev. **133**, B613 (1964).

mass-yield curves for fission induced by bombardment of Th^{232} with alpha particles of three different energies, and have indicated how these curves might be decomposed into a symmetric component and an asymmetric component. From their data we estimate that there is 8.9, 13.1, 17.7% symmetric fission at the three bombarding energies 22.1, 25.7, and 29.5 MeV, respectively. We make the assumption that the probability for light-charged-particle emission from either the symmetric or the asymmetric mode does not vary with excitation energy. From our data on the variation of the observed probability and from the above values for the fraction of symmetric fission we have calculated that the probability for light-charged-particle emission is 1.7×10^{-3} for the asymmetric mode and 6.4×10^{-3} for symmetric mode. If these values are the same for thermal-neutron-induced fission of U^{235}, the peak-to-valley ratio for fission of this system accompanied by light-charged-particle emission should be about 160, in reasonable agreement with the data of Schmitt, Neiler, Walter, and Chetham-Strode.[28] It would be interesting to measure the yield of light charged particles as a function of mass division at an excitation energy such that symmetric fission is reasonably probable.

The drop in yield of light charged particles from zero to 6.5 MeV may be the result of a barrier penetration phenomenon, inasmuch as spontaneous fission is a subbarrier process, whereas thermal-neutron-induced fission is superbarrier.

ACKNOWLEDGMENTS

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APPENDIX I

Many measurements of the probability of light-charged-particle emission have been made relative to the fissioning systems U^{235} plus thermal neutrons and Cf^{252}. We have therefore made a compilation of the results obtained for these two fissioning systems and have also added a measurement of our own, described below, for Cf^{252}. (See Table V.) In our compilation we have required that estimates of the standard error and the low-energy cutoff of the detected spectrum be given.

A chi-square test on the U^{235} data indicates a vanishingly small probability that all of the results should be included. The values measured by Hill and by Marshall are 7 and 9 standard deviations away from the average. Rejecting these we obtain a value of 497 ± 41 (where the error represents the weighted root-mean-square of the deviations). The value of chi-square for this set of numbers is still rather high, indicating that the experimenters have been unrealistic in estimating their errors. There seems to be no justification, however, for rejecting any other data, nor does such rejection make an appreciable difference in the calculated average.

In our measurements with Cf^{252}, the rate of alpha-particle emission in fission and the rate of spontaneous fission were determined with the same detector geometry. For the alpha-particle measurements, the counter telescope was covered with 7.6 mg/cm² of aluminum to stop fission fragments and alpha particles from natural decay. The alpha particles were distinguished from singly charged particles but not from other helium isotopes by their respective ΔE–$(E+\Delta E)$ characteristics. For the fission-fragment measurements, the cover foils were removed and a lower level discriminator was set to prevent the natural decay alpha particles from being counted. A negligible fraction of the fission fragments fell below this discriminator level.

TABLE V. Number of binary fissions per light charged particle for U^{235} plus thermal neutrons and for the spontaneous fission of Cf^{252}.

Ratio	Method	References
	U^{235} plus thermal neutrons	
518 ± 13	Counter	a
512 ± 14	Counter	a
490 ± 20	Counter	a
449 ± 30	Counter	b
220 ± 33	Counter	c
505 ± 50	Counter	d
594 ± 65[e]	Counter	f
401 ± 50[e]	Emulsion	g
230 ± 26	Emulsion	h
340 ± 40	Emulsion	i
333 ± 111	Emulsion	j
	Cf^{252}	
299 ± 18	Counter	b
266 ± 17[e]	Counter	k
397 ± 42[e]	Counter	l
$309+20$[e]	Counter	m
289 ± 16	Counter	n
280 ± 25	Emulsion	o

[a] I. G. Schröder, A. J. Deruytter, and J. A. Moore, Phys. Rev. **137**, B519 (1965).
[b] Reference 12.
[c] David L. Hill, Phys. Rev. **87**, 1049 (1952).
[d] K. W. Allen and J. T. Dewan, Phys. Rev. **80**, 181 (1950).
[e] These data have been corrected from the values by authors to account for the particles below the level of the lower discriminator. The corrections are based on the energy spectrum given by Nobles for Cf^{252} (Ref. 12).
[f] Reference 20.
[g] E. W. Titterton, Nature **168**, 590 (1951).
[h] L. Marshall, Phys. Rev. **75**, 1339 (1949).
[i] L. L. Green and D. L. Livesey, Phil. Trans. Roy. Soc. (London) **241A**, 323 (1948).
[j] Tsien San-Tsiang, Ho Zah-Wei, R. Chastel, and L. Vigneron, Compt. Rend. **224**, 272 (1947).
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[l] M. Luis Muga, Harry R. Bowman, and Stanley G. Thompson, Phys. Rev. **121**, 270 (1961).
[m] Reference 4.
[n] This work.
[o] E. W. Titterton and T. A. Brinkley, Nature **187**, 228 (1960).

[28] H. W. Schmitt, J. H. Neiler, F. J. Walter, and A. Chetham-Strode, Phys. Rev. Letters **9**, 427 (1962).

In a run lasting 45 h we detected 8044 alpha particles (including a few heavier particles[8]) that deposited an an energy greater than 3 MeV in the detector system (equivalent to 7.5 MeV incident on the aluminum foil). Of these, 60, which produce a single peak appropriate to 6.1-MeV alpha particles, were attributed to a Cf^{252} contamination either of the detectors or of the cover foil facing the ΔE detector. From the spectrum reported by Nobles[12] we estimate that $94\pm5\%$ of the so-called long-range particles ($E\geq1.5$ MeV) have energies greater than 7.5 MeV. During an 8-min run with the cover foil removed, we detected 15 428 fission fragments, all assumed to originate from binary fission. From these numbers we calculate a probability for particle emission in the fission of Cf^{252} of $(3.26\pm0.18)\times10^{-3}$. We increase this number by 1.06 ± 0.01 to take into account the tritons[4,5,7] which were not included in our alpha-particle measurement, and obtain $(3.46\pm0.19)\times10^{-3}$ for the probability for light-charged-particle emission, or 289 ± 16 binary fissions per light charged particle.

The weighted average for all the values given in Table V is (292.2 ± 8.1). Elimination of the datum with the largest uncertainty, however, increases from 0.046 to 0.35 the probability that a random sample gives no better fit as determined by the chi-square test. The weighted average of the remaining five values and the value that we adopt is then 288 ± 8, where, in view of the favorable chi-square test we determine the over-all uncertainty from the reciprocal of the square root of the sum of the individual weights. We obtain from this value the probability for light-charged-particle emission in the spontaneous fission of Cf^{252}, $Y_{LC}'=(3.47\pm0.10)\times10^{-3}$, and the alpha-particle emission probability, $Y_\alpha'=(3.27\pm0.10)\times10^{-3}$.

APPENDIX II

TABLE VI. Number of binary fissions per light charged particle for various systems.

System	Ratio	References
Initial compound nucleus U^{234}		
U^{233}+thermal neutrons	414± 26	a
	405±30	b
	428±40	c
	407±39	d
	412±16	Average of four previous values
U^{233}+0.33-MeV neutrons	335±35	e
U^{233}+0.69-MeV neutrons	443±51	e
U^{233}+1.17-MeV neutrons	463±55	e
U^{233}+1.99-MeV neutrons	392±58	e
Initial compound nucleus U^{236}		
U^{235}+thermal neutrons	496± 41	See text
U^{235}+0.33-MeV neutrons	496± 76	e
U^{235}+1-MeV neutrons	534± 35	a
U^{235}+1.17-MeV neutrons	605± 83	e
U^{235}+2.5-MeV neutrons	397± 64	e
	522± 62	f
U^{235}+3.-MeV neutrons	596± 65	g
U^{235}+14-MeV neutrons	1350±190	h
	496± 64	f
Th^{232}+29.5-MeV alpha particles	510± 52	i
Th^{232}+42.0-MeV alpha particles	368± 38	i
Initial compound nucleus U^{239}		
U^{238}+2.5-MeV neutrons	2095±553	e
	600± 84	j
U^{238}+14-MeV neutrons	1000±160	k
	1050±100	l
Ratio: 14-MeV neutrons/ 2.5-MeV neutrons	1.21±0.17	f
Initial compound nucleus Np^{239}		
U^{238}+10.5-MeV protons	602±73	i
U^{238}+17.5-MeV protons	476±45	m
Initial compound nucleus Pu^{240}		
Pu^{240} spontaneous fission	269± 28	n
	314± 20	a
	400± 60	o
	303± 37	Average of three previous values
Pu^{239}+thermal neutrons	411± 26	a
	445± 35	b
	420± 41	d
	477± 48	c
	430+ 20	Average of four previous values
Pu^{239}+0.33-MeV neutrons	410± 43	e
Pu^{239}+0.69-MeV neutrons	478± 57	e
Pu^{239}+1-MeV neutrons	403± 22	a
Pu^{239}+1.99-MeV neutrons	400±102	e
Initial compound nucleus Pu^{242}		
Pu^{242} spontaneous fission	365±29	a
Pu^{241}+thermal neutron	440±28	a
	370±36	c
U^{238}+29.5-MeV alpha particles	391±44	i
U^{238}+42-MeV alpha particles	284±23	i

[a] Reference 12.
[b] K. W. Allen and J. T. Dewan, Phys. Rev. **80**, 181 (1950).
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[e] M. F. Netter, H. Faraggi, A. Garin-Bonnet, M. J. Julien, C. Corge, and J. Turkiewicz, in *Proceedings of the Second United Nations Conference on the Peaceful Uses of Atomic Energy* (United Nations, Geneva, 1958), Vol. 15, p. 418, *P*/1188.
[f] Reference 15.
[g] Reference 20. We have used the ratio of the measurements made by these workers for thermal and 3-MeV neutrons together with the average value for thermal neutrons given in Appendix I.
[h] Reference 13.
[i] Reference 14.
[j] Z. I. Solov'eva, At. Energ. (USSR) **8**, 137 (1960).
[k] N. A. Perfilov and Z. I. Solov'eva, At. Energ. (USSR) **5**, 175 (1958).
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[m] Reference 17.
[n] Reference 11.
[o] N. A. Perfilov, Z. I. Solov'eva, R. A. Filov, and G. I. Khlebnikov, in *Physics of Nuclear Fission*, edited by N. A. Perfilov and V. P. Eismont, Atomic Energy Commission Report No. AEC-tr-6205, (1965) p. 125.