Long-Range Particles from Nuclear Fission*

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A measurement of the probability of emission of long-range particles, which are known to be predominantly alpha particles, made with a multiple ionization chamber gave the following results for spontaneous fission: Cf^{252}, 299±18; Cm^{242}, 257±17; Cm^{244}, 314±20; Pu^{240}, 314±20; and Pu^{242}, 365±29 fissions per long-range particle. The results for thermal neutron-induced fission of the following compound nuclei were: Pu^{240}, 411±26; Pu^{242}, 440±28; U^{236}, 449±30; and U^{234}, 414±26 fissions per long-range particle and for 1-Mev neutron-induced fission the results were: Pu^{240}, 403±25; U^{236}, 534±35; U^{235}, 466±30 fissions per long-range particle. These data indicate a trend of increasing probability of emission of long-range particles with increasing Z^2/A values and decreasing probability with increasing excitation energy in a given nuclide. Present explanations of these apparently contradictory results seem inadequate.

The energy spectra of long-range particles from spontaneous fission of Cf^{252} and thermal neutron-induced fission of U^{235} were observed with a CsI crystal scintillation apparatus and were found to be identical within the accuracy of observations, showing slightly askew bell-shaped distributions with maxima at 17±1 Mev and end-point energies of 29±1 Mev.

INTRODUCTION

NUMEROUS studies have established[1,2] that long-range particles are emitted by most fissioning nuclei with a frequency of about 1:250 to 1:500 ordinary fission events and that most of these particles have the e/m and range-energy characteristics of alpha particles. It has also been established that their energy and angular distributions are consistent with the hypothesis that they are emitted in the region of the neck at the time of scission.

To provide additional information on the emission of these particles, this paper reports results of a systematic study of the probability of emission of long-range particles as a function of Z^2/A of spontaneously fissioning nuclei and as a function of the excitation energy inducing fission. A multiple ionization-chamber apparatus which required a threefold coincidence between fission fragments and a long-range particle was used to observe this ratio for several nuclides undergoing either spontaneous fission, slow neutron-induced fission or 1-Mev neutron-induced fission. Also reported are the results of a CsI crystal-scintillation-spectrometer study of the energy spectra of the long-range particles from the spontaneous fission of Cf^{252} and thermal neutron-induced fission of U^{235}.

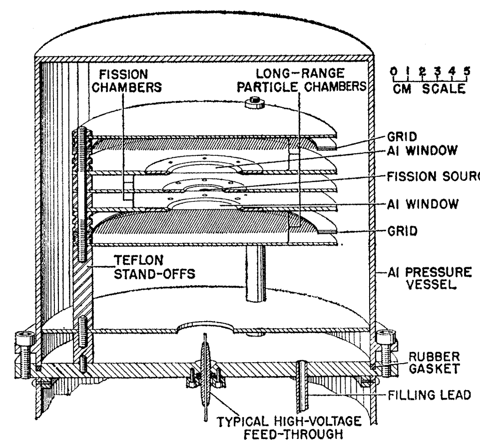

FIG. 1. Ionization chamber apparatus used in the measurement of the probability of emission of long-range particles in fission.

APPARATUS AND EXPERIMENTAL PROCEDURES

The apparatus used in the measurement of the probability of long-range particle emission in fission, that is of the ternary-to-binary fission ratio, consisted of four parallel-plate ionization chambers operating in an argon plus 5% CO_2 gas mixture. The arrangement of the ionization chambers is shown in Fig. 1. The fission source was deposited on a 100-μg/cm^2 nickel foil, which was mounted on a centrally located electrode. The electrodes on either side of the central electrode acted as electron collectors for the fission chambers and also contained 3.2-mg/cm^2 aluminum windows which allowed the long-range particles to enter the outer ionization chambers. The filling gas pressure was adjusted so that the stopping power of the fission chamber plus that of the aluminum window was just sufficient to stop fission fragments and alpha particles from radioactive decay. Grids were employed in the outer chambers to insure that long-range particles which were stopped in the chambers would give pulses whose heights were proportional to their energy loss. This simplified the task of extrapolating to the unobserved low-energy portion of the long-range particle-energy spectrum. This unobserved portion was comprised of particles with insufficient range to enter the outer chambers. The fission-chamber collector electrodes also served as ground electrodes for the outer chambers; hence, these electrodes

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† Present address: Lockheed Missiles and Space Company, Palo Alto, California.
[1] W. J. Whitehouse, in *Progress in Nuclear Physics* (Academic Press, Inc., New York, 1952), Vol. 2, pp. 159–163.
[2] N. A. Perfilov, Yu. F. Romanov, and Z. I. Solov'eva, Soviet Phys.—Uspekhi **3**, 542–549 (1961).

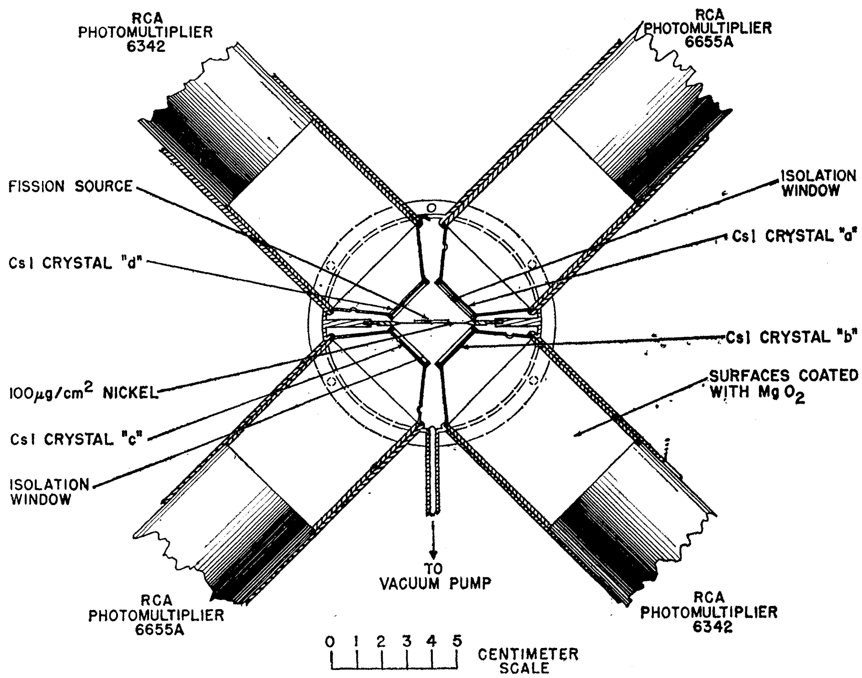

FIG. 2. CsI crystal scintillation-spectrometer apparatus used in the measurements of the energy spectrum of long-range particles from fission. Crystals "*a*" and "*c*" are fission detectors and "*b*" and "*d*" long-range particle detectors. The thickness of the fission source is shown greatly exaggerated.

were shunted to ground with a resistor whose value was chosen so as to permit the fission pulse to be picked up and at the same time not allow a large enough voltage change to transmit the pulse to the signal plates of the outer chambers.

The mode of operation of the apparatus was such that a twofold coincidence between pulses from the fission chambers was recorded as a binary-fission event, and a threefold coincidence between pulses from the fission chambers and a pulse from a long-range particle chamber was recorded as a ternary fission. To yield the true ratio of ternary-to-binary fission events or the probability of long-range particle emission, the data were corrected for geometry, for accidental coincidences, for the unobserved low-energy portion of the spectrum, and for self-absorption in the source, account being taken of the fact that the long-range particles are emitted at approximately 90° to the direction of the fragments. Standard electronic circuits and coincidence techniques were employed in the collection of the twofold and threefold coincidence data and for simultaneous collection of accidental coincidence background data.

For measurements of 1-Mev neutron-induced fission, one of the long-range particle chambers was removed and the remaining electrode assembly moved closer to the end wall of the chamber to decrease the mean distance between the fission sample and neutron source. Measurements taken with a Cf^{252} spontaneous fission source normalized the data taken with this set up to those taken with the double-chamber arrangement.

Most of the fission sources were prepared by the techniques of electrospraying,[3] vacuum evaporation, and painting. The Cf^{252} source was prepared by autodeposition[4] from an intense parent source. Deposits were made on 100-μg/cm^2 nickel foil[5,6] which in some cases was reinforced by a backing of 20-mesh, 93%-transparent, "electromesh" nickel screen. It was necessary to cover some of the highly alpha-active sources with 100-μg/cm^2 nickel foil to prevent the source material from spreading throughout the chamber. Table I lists the principal characteristics of the fission sources used in the work reported in this paper. The isotopic compositions of the sources were such that 95% or more of the fissions observed were from the isotope listed.

The thermal-neutron-induced fission measurements were made with the external neutron beam from the south thermal column of the Los Alamos Homogeneous Reactor and the 1-Mev measurements were made with neutrons from the $T(p,n)$ reaction in the large Van de Graaff accelerator at Los Alamos.

The CsI crystal scintillation apparatus used to study the energy spectrum of the long-range particles from fission is illustrated in Fig. 2. This apparatus consisted of a fission source deposited on a 100-μg/cm^2 nickel foil symmetrically positioned with respect to two sets of diametrically opposed thallium-activated CsI crystals, which were viewed by photomultiplier tubes through MgO_2-surfaced vacuum light pipes. Two of the crystals, "*b*" and "*d*" in Fig. 2, were used as long-range particle detectors. These crystals were 2 cm in diameter and 0.41 mm in thickness; this thickness was sufficient to stop

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[4] Process employs evaporation from local heating by fission fragments: B. V. Ershler and F. S. Lapteva, J. Nuclear Energy 4, 471 (1957).

[5] Chromium Corporation of America, Waterbury, Connecticut.

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TABLE I. Principal characteristics of the sources used in long-range particles from fission measurements.

Fission source	Method of preparation	Amount of active material	Spontaneous fission rate (fissions/min)
Cf^{252} [a]	Autodeposition	$\approx 10^{-3}$ μg	3×10^4
Cm^{242}	Electrospray	≈ 0.06 μg 5×10^8 α/min	17
Cm^{244}	Electrospray	≈ 2 μg 5.4×10^8 α/min	390
Pu^{240}	Electrospray	0.18 mg	5.1
Pu^{241}-Pu^{242} [b]	Electrospray	0.29 mg	12.4
Pu^{239}	Vacuum evaporation	0.123 mg	
U^{233} [c]	Vacuum evaporation	1.0 mg	
U^{234}	Brush painting	0.890 mg	
U^{235}	Vacuum evaporation	0.860 mg	

[a] Source used for calibration of the apparatus throughout the experiments.
[b] Pu^{242}, 88.5%; Pu^{241}, 7.5%. Source used in spontaneous and thermal neutron-induced fission measurements.
[c] Freshly separated sample used in order to eliminate high-energy alpha-producing daughters.

30-Mev alpha particles at normal incidence. Crystals "*a*" and "*c*" were of the same diameter as "*b*" and "*d*" and were used in twofold coincidence as fission detectors. Their 0.076-mm thickness was a compromise between undesirable sensitivity to gamma radiation and mechanical strength. These fission detectors were covered with 100-μg/cm^2 nickel-foil light shields to prevent "crosstalk" with the long-range particle crystals. The energy resolution of the long-range particle counters was about 11% for 6.1-Mev Cf^{252} alphas and 6% for 8.78-Mev alphas from Po^{212}.

The mode of operation of the apparatus was as follows: A twofold coincidence between pulses from the fission detectors established the occurrence of a fission event. A threefold coincidence with the fission pulses and a third pulse from either of the long-range particle detectors opened a gate on a 400-channel pulse-height analyzer which analyzed the third pulse and addressed the information to one 200-channel memory bank of the analyzer. In an analogous manner pulses were fed into a second coincidence circuit at random time with respect to the fission pulses by a pulser whose repetition rate was set the same as the average fission-coincidence pulse rate. Chance pulses in the long-range particle channels in coincidence with these random pulses were analyzed and addressed to the second half of the memory; thus a background distribution was secured which was later subtracted from the pulse-height distribution stored in the first half of the memory.

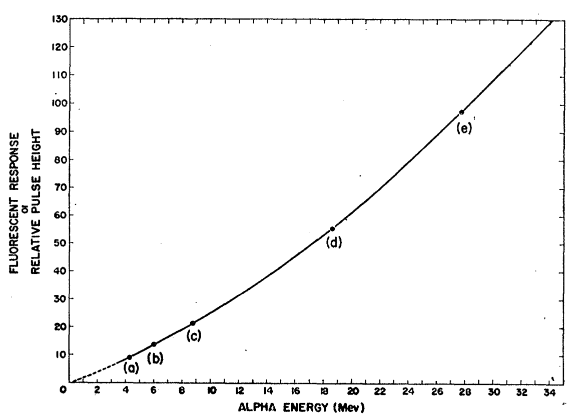

FIG. 3. Fluorescent response of CsI crystals as a function of alpha-particle energy. (a) U^{235} alpha particles; (b) Cf^{252} alpha particles; (c) Po^{212} alpha particles; (d) and (e) alpha particles from cyclotron.

The fluorescent response of CsI crystals to alpha particles was determined by the use of alpha particles both from natural radioactivity and from the Los Alamos variable-energy cyclotron. Figure 3 shows the results of these fluorescent responses (relative pulse height) versus alpha particle energy measurements; these results are in good agreement with those reported by Quinton *et al.*[7] The experimental points in the figure represent a composite of the data from crystals "*b*" and "*d*".

RESULTS: MEASUREMENTS OF LONG-RANGE PARTICLE-EMISSION PROBABILITY

The results of the measurements of the probability of the emission of long-range particles are given in Table II. A plot of the number of long-range particles per fission as a function of Z^2/A and of the parameter X is given in Fig. 4. The fissionability parameter, X, can be written as $X=(Z^2/A)/(Z^2/A)_0$, where $(Z^2/A)_0 \approx 50.13$ is the limiting value for a spherical nucleus which is just unstable with respect to small distortions.[8]

Table III lists the relative probability of emission of long-range particles from a given nuclide as a function of excitation energy. The excitation energy is the sum of the neutron kinetic energy and its binding energy.

TABLE II. Results of measurements of the probability of long-range particle emission.

Nuclide	Type fission	Z^2/A	Fissions per long-range particle
Cf^{252}	Spontaneous	38.11	299±18
Cm^{242}	Spontaneous	38.08	257±17
Cm^{244}	Spontaneous	37.77	314±20
Pu^{240}	Spontaneous	36.82	314±20
Pu^{240}	$Pu^{239}+n_{Th}$	36.82	411±26
Pu^{240}	$Pu^{239}+n_{1\ Mev}$	36.82	403±25
Pu^{242}	Spontaneous	36.51	365±29
Pu^{242}	$Pu^{241}+n_{Th}$	36.51	440±28
U^{236}	$U^{235}+n_{Th}$	35.86	499±30
U^{236}	$U^{235}+n_{1\ Mev}$	35.86	534±35
U^{235}	$U^{234}+n_{1\ Mev}$	36.02	466±30
U^{234}	$U^{233}+n_{Th}$	36.17	414±26

[7] A. R. Quinton, C. E. Anderson, and W. J. Knox, Phys. Rev. **115**, 886 (1959).

[8] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).

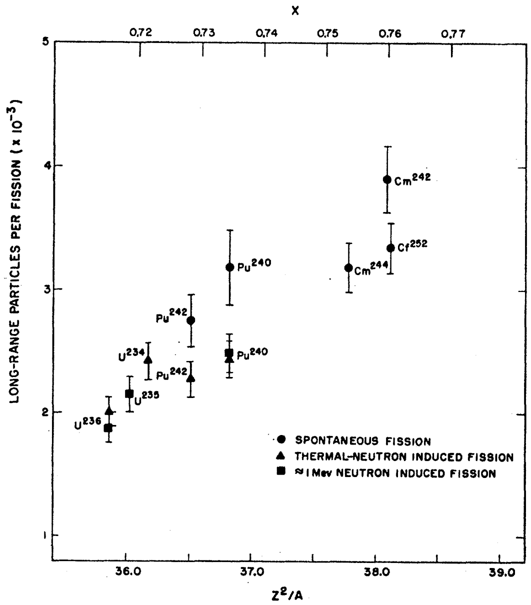

FIG. 4. The number of long-range particles per fission as a function of Z^2/A and X.

TABLE III. Variation of relative probability of long-range particle emission with excitation energy.

Nuclide	Type fission	Neutron energy (Mev)	Excitation (Mev)	Relative probability of long-range particle emission
Pu^{240}	Spontaneous	···	0	1
Pu^{240}	$Pu^{238}+n$	Thermal	6.4	0.765 ± 0.086
Pu^{240}	$Pu^{239}+n$	1.0	7.4	0.780 ± 0.090
Pu^{242}	Spontaneous	···	0	1
Pu^{242}	$Pu^{241}+n$	Thermal	6.3	0.829 ± 0.084
U^{236}	$U^{235}+n$	Thermal	6.5	1
U^{236}	$U^{235}+n$	1.0	7.5	0.934 ± 0.082

RESULTS: LONG-RANGE PARTICLE ENERGY-SPECTRUM MEASUREMENTS

The threefold-coincidence data from the Cf^{252} source gave a distribution of counts versus pulse height which was converted to counts-per-energy-interval versus energy by the use of the fluorescent-response data of Fig. 3. The results of the observation are shown in the upper curve of Fig. 5. No data points are given below 7.55 Mev because the data below this energy were taken in a separate run employing an expanded energy scale (i.e., higher gain) in order to observe the region in greater detail. These data were fitted to the main distribution at 7.5 Mev.

No significant data were obtained in the vicinity of 6.1-Mev energy because of the large statistical uncertainty of the results in this region. This statistical uncertainty was due to the high background of accidental threefold-coincidence pulses caused by the alpha particles from radioactive decay. Figure 5 also shows the energy distribution observed for long-range particles from thermal-neutron-induced fission of U^{235}, i.e., from the compound nucleus U^{236}.

DISCUSSION OF RESULTS

In Fig. 4 the number of long-range particles per fission increases with increasing Z^2/A values, and Table III shows that the number of long-range particles decreases with increasing excitation energy of the fissioning nucleus. This increasing excitation energy is associated with increasing angular momentum because of neutron-orbital-angular momentum in addition to the spins of the neutron and the target nucleus. The resulting increase in the fission threshold compared to the threshold of the same nucleus with zero spin, which is the case for the spontaneous fissions being considered, is the order of 1 Mev or less[9]; whereas the excitation from neutron capture is the binding energy of the neutron, approximately 6 Mev, plus the kinetic energy of the neutron. Hence, one would expect the excitation from neutron capture to predominate in determining the number of long-range particles emitted.

In Fig. 4 the number of long-range particles per fission observed for spontaneous fission of Cm^{242} is about two standard deviations off from the value expected from an interpolation of the values observed for the neighboring nuclides Cf^{252} and Cm^{244}. There is no apparent reason for the anomalously high Cm^{242} value. It is interesting to note that Perfilov *et al.*[10] have recently reported a measurement giving $(3.3\pm0.39)\times10^{-3}$ alphas per fission for Cm^{242}, a value which fits the systematics of the data presented in Fig. 4. However, a much lower value has been obtained by Henderson *et al.*[11]

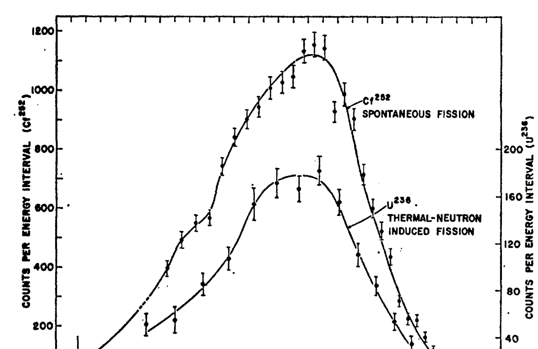

FIG. 5. Energy distribution of long-range particles from spontaneous fission of Cf^{252} and from thermal-neutron-induced fission of U^{235}, i.e., the compound nucleus U^{236}. The ordinates are normalized so that the area under each curve is proportional to the number of long-range particles per fission.

[9] J. Griffin, in *Proceedings of the International Conference on Nuclear Structure, Kingston* (University of Toronto Press, Toronto, 1960), Chap. 9.

[10] N. A. Perfilov, Z. I. Solov'eva, R. A. Filov, and G. I. Khlebnikov, Soviet Phys.—Doklady **6**, 57 (1961).

[11] D. J. Henderson, H. Diamond, and T. H. Braid, Bull. Am. Phys. Soc. **6**, 418 (1961).

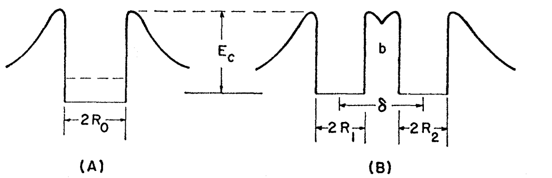

FIG. 6. "Sudden approximation" theory potential distribution. (A) shows the original potential well, $R_0=r_0A^{\frac{1}{3}}$ cm, $E_c\approx 28$ Mev is the height of the Coulomb barrier, and the broken line in the well indicates the alpha-particle binding energy. (B) shows the potential distribution immediately after scission, $R_1\approx R_2\approx r_0(A/2)^{\frac{1}{3}}$ cm, and δ is the breaking distance (i.e., the distance between the fragment mass centers at scission).

The results of the energy-spectrum observations given in Fig. 5 are in general agreement with work previously reported.[12–15] Both of the distributions have maxima in the vicinity of 17 Mev, end-point energies of 29 ± 1 Mev, and full widths at half-maximum of about 11 Mev. However, Muga *et al.*[16] report a peak at 19 ± 1 Mev and an end-point energy of 34 Mev for the energy spectrum of long-range particles from spontaneous fission of Cf^{252}; no reason for this discrepancy is apparent. The similarity of the shape and of the end-point energies of the curves in Fig. 5 strongly suggests that the same basic mechanism is involved in both cases. This conclusion is consistent with the hypothesis which has the third particle produced at scission in the region of the neck with low-kinetic energy and the three fragments then separating under their mutual-electrostatic repulsion.

The curve representing the energy distribution of particles from the spontaneous fission of Cf^{252} is drawn with a shoulder on the low-energy side of the distribution. This structure is suggested by the data points and is given credence by the existence of the triton group reported by Watson[17] and Wegner.[18] According to Wegner these particles have an energy distribution with a peak at 8.5 ± 1.0 Mev, an end-point at 16 ± 1 Mev, and a probability of occurrence about 6% that of alpha particles. These data are consistent with the structure in Fig. 5.

MECHANISM FOR THE EMISSION OF LONG-RANGE PARTICLES

In his work on binary and ternary fission of heavy elements,[19] Tsien considers a simplified scheme of tripartition which envisions the nuclear matter at scission as having the configuration of three spheres in contact. The three spheres are considered to be at rest and then after separation are accelerated by their mutual electrostatic repulsion. The final energy of the light fragment depends, among other things, upon the initial distance of its center of mass from the center line of the two heavy fragments.

This theory provides a prediction for the maximum energy of long-range particles which is in good agreement with the observed end-point energy, and also provides a qualitative explanation for the observed shape of the energy distribution as well as for the observed average angle of emission of the third particle with respect to the direction of the heavy fragments.

Tsien speculates that there is a uniform probability for the production of third particles in fission and explains the observed decrease in the number of long-range particles with increasing excitation energy by supposing that the average value of the separation distance of the center of mass of the light fragment from the center line of the heavy fragments decreases with increasing excitation of the compound nucleus, thus yielding a decreased number of high-energy, i.e., long-range particles. However, an analogous line of reasoning leads to the expectation of decreasing average separation distances with increasing Z^2/A values and hence to decreased probability of emission of long-range particles, which is contrary to observation.

The barrier penetration model[20,21] which is used to describe nuclear-particle emission does not appear to be applicable to long-range particle emission in fission for the following reason. The semiclassical theory of radioactive alpha-particle emission by barrier penetration involves the rate at which the alpha particles hit the barrier multiplied by the transparency of the barrier. This theory predicts half-lives for the process of the order of thousands of years; but the particles emitted in fission, most of which are alphas, are emitted at the time of scission, as is evidenced by their angular distribution. It is not obvious what time interval, Δt, is relevant to the process; however, Δt probably lies somewhere between the mean lifetime of the compound nucleus ($\approx 10^{-14}$ sec), and the time required for a nucleus to pass from the Bohr-Wheeler saddle-point configuration to the scission or fragment-separation point. This latter time is estimated to be of the order of several nuclear-transit times (i.e., $\Delta t\approx 10^{-21}$ sec); hence, 10^{-21} sec $<\Delta t<10^{-14}$ sec. The transparency of the barrier in the region of the neck of a nucleus near the scission point is expected to be about the same as that of a normal nucleus, and the alpha particle's "free energy" not greatly different from that in a normal nucleus; hence, there is not enough time for alpha particle emission by barrier penetration by many orders of magnitude.

The possibility that the particles are produced by nuclear evaporation from either the original nucleus or

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15 N. A. Perfilov, "Physics of Fission" in Supplement No. 1, Soviet Journal of Atomic Energy (Pergamon Press, New York, 1957), p. 98.
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17 J. C. Watson, Phys. Rev. **121**, 230 (1961).
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20 G. Gamow, Z. Physik **51**, 204 (1928).
21 R. W. Gurney and E. U. Condon, Nature **122**, 439 (1928).

from the fragments after scission can be ruled out, because the former process would lead to an isotropic-angular distribution and the latter to a distribution tending to peak in the direction of motion of the fragments, whereas the long-range particles are actually strongly peaked at $\approx 90°$ to the direction of the fragments. However, the possibility that the density and temperature of the nuclear matter in the neck of a nucleus just before scission may be such as to be favorable for formation and evaporation of an alpha particle, for instance, cannot be excluded by such *a priori* angular-distribution arguments.

Halpern has proposed a theory for the production mechanism for the long-range particles based on the "sudden approximation," or nonadiabatic nuclear-potential change.[22] In this theory the alpha particle is originally held in a potential well, shown at (A), Fig. 6. At scission the potential changes to (B), Fig. 6. This change is assumed to occur in a time which is short compared to the alpha particle's nuclear-transit time; a calculation is made of the probability that the alpha particle will be trapped in region b after scission. Halpern's preliminary calculations indicate results of the correct order of magnitude for the probability of emission of alpha particles. However, the theory in its present state of development does not appear to involve the proper parameters to allow a prediction of the variation of the probability of particle emission for different nuclides and excitation energies.

Fuller has examined in greater detail a similar theory on the effect of nonadiabatic potential change on neutron production in fission.[23] He found that with a reasonable choice of parameters the theory gave a prediction in general agreement with the experimental work of Bowman *et al.*[24] on the number of "scission neutrons" produced in spontaneous fission of Cf^{252}. A further development of the "sudden approximation" with reference to the emission of charged particles in fission might be of interest.

From this discussion no satisfactory explanation is apparent for both the variation from nuclide to nuclide of the probability of emission of long-range particles and the variation with excitation energy in a given nuclide. Indeed, increasing probability of emission with increasing Z^2/A appears inconsistent with decreasing probability of emission with increasing excitation energy when viewed from the standpoint of obvious correlations such as, for example, the height of the fission barrier for spontaneous fission and the energy in excess of the barrier for induced fission.

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