Emission of Alpha Particles in the Fission Process

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The angular distribution with respect to the incident neutrons of the long-range alpha particles emitted in the fission of U^{238} by 14-MeV neutrons is investigated using the emulsion technique. The shape of this distribution suggests that the alpha emission is a compound nucleus evaporation process. The energy spectrum of alphas emitted in the fission of U^{235} is also reasonably accounted for by a new mechanism proposed here which assumes that the emission of alphas takes place from the neck of the distorted compound nucleus with a charge polarization along the fragment axis.

THE emission of alpha particles in fission has been studied by many workers and the earlier work is summarized in the comprehensive review by Hyde.[1] Perfilov[2] has recently reviewed the various aspects of this process, or "ternary" fission as it is called, and it can be said that no existing theory of the process is able to explain even qualitatively all the observed features. As ternary fission of heavy nuclei induced by energetic projectiles is expected to give more information about the nature of the process, because of high angular momentum imparted to the nucleus, a study of the ternary fission of U^{238} induced by 14-MeV neutrons has been made here using the nuclear emulsion technique. These experimental findings and other published results can be reasonably accounted for on the mechanism of ternary fission proposed here.

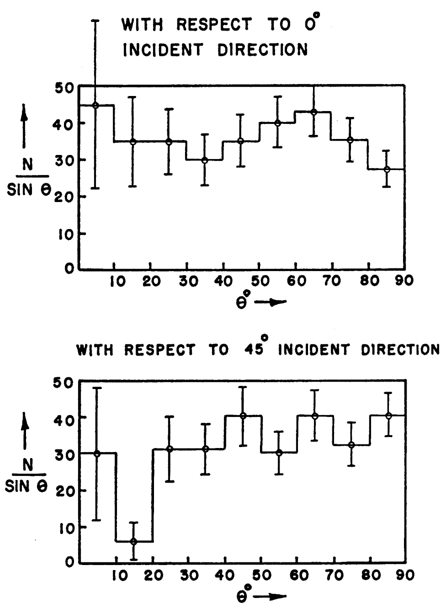

Fig. 1. Angular distributions of natural alpha particles from natural uranium.

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[2] N. A. Perfilov, Yu. F. Romanov, and Z. I. Solov'eva, Soviet Phys.—Uspekhi 3, 542 (1961).

EXPERIMENTAL PROCEDURE AND RESULTS

Ilford C2 100 μ plates were loaded with natural uranium using a 5% aqueous solution of uranium nitrate. The plates were immersed in the solution for about 30 min and afterwards dried and exposed to a beam of 14-MeV neutrons produced in a cascade generator by the (d,t) reaction. The plates were placed at a distance of 25 cm from the tritium target in such a way that the incident neutron beam made angles of 10° and 45° with them, respectively. After processing, the plates were subjected to scanning for binary and ternary fission events, using a Leitz microscope with 800 magnification. For measurements a magnification of 2000 was used for higher accuracy. Measurements could be made with an accuracy of $\pm 0.5\ \mu$ in range and $\pm 5°$ in angles.

About 180 events having long-range alphas associated with the fission fragments were obtained. Total range measurements could not be made in all cases as some of the alphas were either escaping into the air or ending in the glass. These were, however, included for the angular distribution studies. In order to check that all angles were scanned with equal efficiency in both the 10° and 45° plates, the angular distribution of alphas from natural uranium obtained with respect to the 0° and 45° reference directions were found to be isotropic within statistics in both cases (Fig. 1). The ternary fission events were analyzed only in the 45° plate. The binary fission events were analyzed in both the 10° and 45° plates.

The angular distribution of the alphas with respect to the incident beam direction shows a forward-backward peaked distribution, symmetric about 90° (Fig. 2). The angular distribution of the binary fission fragments with respect to the incident neutron direction is shown in Fig. 3. Our measurements on the angular distribution of binary fission fragments show an anisotropy $N(0°)/N(90°) \sim 2$. The angular distribution of binary fission

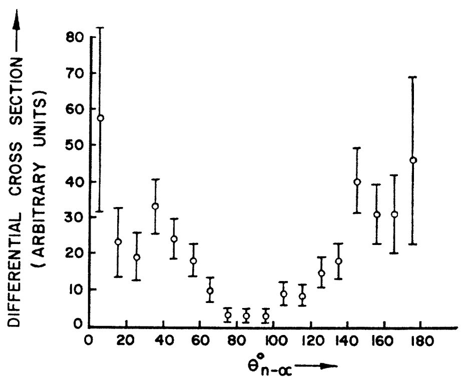


FIG. 2. Angular distribution of long-range alpha particles with respect to the incident neutron direction.

fragments has also been measured by other workers.[3–6] Their results, obtained with better statistics, however, show a smaller ratio (1.3 to 1.4).

The angular distribution of the fission fragments for ternary fission events is given in Fig. 4.

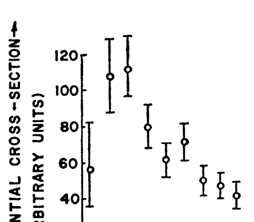
Figure 5 shows the energy distribution of the long-range alphas obtained from our measurements for the case of 14-MeV neutron fission of U^{238}. The continuous line shows the alpha energy distribution for the case of thermal neutron fission of U^{235} as obtained by the magnetic analysis of Fulmer and Cohen.[7]

DISCUSSION

Previous investigations[2] have shown that the relative probability of ternary fission compared to binary fission is about 1:300 for thermal neutrons and decreases to about 1:600 and 1:1100 for 2.5- and 14-MeV neutron-induced fission,[8] respectively.

The most probable angle of emission of alpha particles is about 90° to the direction of motion of the

FIG. 3. Angular distribution of binary fission fragments with respect to the incident neutron direction.



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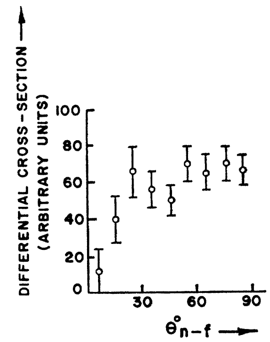
[6] J. E. Brolley, Jr., and W. C. Dickinson, Phys. Rev. **94**, 640 (1954).
[7] C. B. Fulmer and B. L. Cohen, Phys. Rev. **108**, 370 (1957).
[8] N. A. Perfilov and Z. I. Solov'eva, At. Energ. (USSR) **5**, 175 (1958).

FIG. 4. Angular distribution of ternary fission fragments with respect to the incident neutron direction.



fragments. The angular distribution of the alpha particles becomes broader[9] with increasing energy, approaching an isotropic distribution for very high energies ($E \gtrsim 21$ MeV).

The measurements by Dmitriev *et al.*[10] on the kinetic energy distribution of the fission fragments in coincidence with alpha particles show that the sum of the most probable values of the kinetic energies of the fragments and the emitted alpha particles in ternary fission is nearly equal to the most probable value of the kinetic energies of the fragments in binary fission. Further, the probability of ternary fission does not depend on the mass ratio of the fragments. These observations go against the proposal[11,12] that the alpha particles are emitted by a special deformation configuration at the neck. The explanation of the alpha particles being emitted from the moving fission fragments can be ruled out as the angular correlation of the alphas and fragments do not indicate this.

Measurements by Apalin[13] show that lesser number of prompt neutrons are emitted from ternary fission

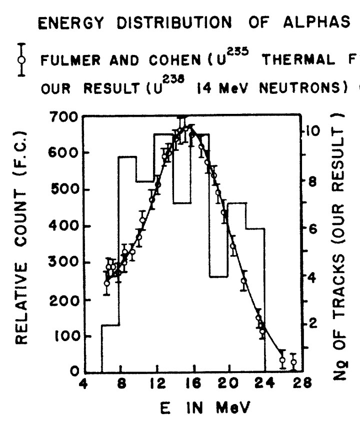


FIG. 5. The energy distribution of the long-range alpha particles.

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events as compared to binary fission, indicating lesser excitation energy of ternary fission fragments. On the statistical theory of fission,[14] the lesser probability of ternary fission as compared to binary fission can be attributed to the decrease in the excitation energy of the system at the scission point. According to Dmitriev *et al.*,[10] this decrease in the excitation energy of the system does not depend much on the mass ratio. The relative probability of ternary fission should, therefore, be independent of the mass ratio as observed.[10] The applicability of the statistical theory of fission to account for these facts may be taken as an indication that alpha emission takes place when the system is in statistical equilibrium.

The following discussion seeks to explain the experimental results on the hypothesis that the alpha particles are evaporated from the excited and deformed nucleus prior to fission. The experimental fact in favor of the proposed hypothesis is the observed angular distribution of the alpha particles (Fig. 2) relative to the incident 14-MeV neutrons, which is characteristic of the particles evaporated from a compound nucleus having a large angular momentum $\bar{I}$. The anisotropy results from the restrictions on the directions of the angular momenta $\bar{l}$ of the emitted particles imposed by the spin distribution of the residual nucleus. For a single-emission process, the anisotropy is given by[15]

$$N(\theta)\simeq[1+\tfrac{1}{2}\alpha_f^2\langle l^2\rangle\langle I^2\rangle\cos^2\theta], \qquad (1)$$

where $\langle I^2\rangle$ and $\langle l^2\rangle$ are the averages of I^2 and l^2 with relative weighting factors $IT(I)$ and $lT(l)$, respectively; $T(I)$ and $T(l)$ are the transmission factors; and $\alpha_f=\hbar^2/2JT$, J being the moment of inertia of the nucleus and T its temperature.

The anisotropy computed from expression (1) is very much smaller than that observed experimentally. This can be qualitatively accounted for on the assumption that in fission induced by high-energy neutrons, the emission of the alpha particles is from the compound nucleus after it has emitted all the possible number of neutrons so that the nucleus after alpha emission has significantly lower average spin values than otherwise. The direction of the alpha-particle angular momentum will, therefore, be more strongly correlated with the original spin, resulting in a larger anisotropy. An alternative explanation could be that the value of the moment of inertia J could be different from that calculated for a spherical nucleus, but the expected change in the value of J will not be sufficient to explain the anisotropy. If alpha emission takes place from the compound nucleus after it has released the extra excitation energy by the emission of neutrons, the alpha spectra for fission induced by 14-MeV neutrons and by thermal neutrons should not be very different. This has been observed experimentally by Perfilov[8] and our measurements also indicate this (Fig. 5).

Fulmer and Cohen[7] have pointed out that the hypothesis completely fails to explain the energy distribution of the emitted long-range alpha particles in the fission of U^{235} induced by thermal neutrons.

According to the theory of statistical evaporation

$$N(E)dE=\text{const}E\sigma_c(E)\omega(E'-E)dE, \qquad (2)$$

where $N(E)$ is the energy distribution of the emitted particles, $\sigma_c(E)$ is the cross section for the inverse process, and $\omega(E'-E)$ is the level density of the residual nucleus. E' is the effective excitation energy of the compound nucleus taking into account the binding energy of the emitted particle.

Introducing the Taylor expansion of $\ln\omega(E'-E)$ up to the first two terms only,

$$N(E)dE=\text{const}E\sigma_c(E)e^{-E/\theta(E')}dE. \qquad (3)$$

The maximum of $N(E)$ lies at energy $E=\theta$ which should be small compared to E' to make the expansion (3) valid. For the case of emission of alpha particles, because of the strong dependence of $\sigma_c(E)$ on E the maximum will be shifted to much higher particle energies than $E=\theta$ and the approximation (3) is no longer true. It will then be necessary to use Eq. (2) directly to compute the spectrum $N(E)$. However, making use of Eq. (3) with a temperature of 1.4 MeV, Fulmer and Cohen[7] find that the variation of computed $\sigma_c(E)$ with E is more rapid than required to explain the alpha spectrum. The computations of $\sigma_c(E)$ were taken from Blatt and Weisskopf[16] for a spherical nucleus. This led them to conclude that alpha particles are not emitted from a compound nucleus process. Our calculations show that the disagreement persists even if one uses the exact expression (3) with the Fermi gas level density dependence given by

$$\omega(E'-E)=Ce^{2a^{\frac{1}{2}}(E'-E)^{1/2}}, \qquad (4)$$

where $a=12$ MeV^{-1} (for $A=231$).

In our calculations $\sigma_c(E)$ was computed assuming $r_0=1.2\times10^{-13}$ cm.

The above anomaly may be the result of not taking into account the complete dynamics of the fissioning nucleus from saddle point to scission and also the increased deformation during alpha emission. After the fissioning nucleus has crossed the saddle point of a particular potential energy curve, the decrease in the potential energy goes to the kinetic energy of the collective motion towards fission, collective deformation of the nuclear surface, and individual particle excitations. Thus, the observed spectrum of alpha particles is an integration of the various spectra emitted in various stages of deformation, each corresponding to an excitation energy E'.

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Taking into account the variation of E', one gets

$$N(E)dE = \text{const}\int_E^\infty EP(E')\sigma_c(E,E') \times\omega(E'-E)dE'dE, \quad (5)$$

where $P(E')$ is the probability of finding the compound nucleus in a stage of excitation E'. This probability is assumed to be given by the Boltzmann factor $\exp(-E'/T_D)$, where T_D is some average temperature of the distribution. The limits of the integration for thermal fission are from E to ∞, since for alpha particles to be emitted with energy E, the contribution is from excitation energies of the compound nucleus from E to ∞.

$\sigma_c(E,E')$ is a function of the energy and deformation of the nucleus. As the excitation energy changes with deformation, $\sigma_c(E,E')$ is a function of E' also. The variation of $\sigma_c(E,E')$ for various stages of deformation can be approximated to an average $\sigma_c^{\text{def}}(E)$ for an average deformation specified by β.

Substituting $\sigma_c^{\text{def}}(E)$ for alpha emission from the deformed nucleus, in (5), one gets

$$N(E)dE = \text{const}E\sigma_c^{\text{def}}(E)e^{-E/T_D}. \quad (6)$$

It is interesting to note that if one assumes a constant-temperature formula for the level density given by $\omega(E'-E) = \text{const}\exp[(E'-E)/T]$, one obtains the same expression as (6). Fulmer and Cohen[7] used the value $\sigma_c^{\text{sph}}(E)$ for the case of a spherical nucleus and found that the observed spectrum is not even in qualitative agreement with the predictions of (6).

From the reciprocity theorem for nuclear reactions, $\sigma_c(E)$ can be approximated with the Coulomb barrier penetration factors for the escape of an alpha particle from the compound nucleus. It is expected that the potential barrier and hence $\sigma_c(E)$ will be different for emission at different stages of deformation. In order to explain the angular correlation of alpha particles with fission fragments, it is necessary to assume that the barrier is lowered at the neck of the fissioning nucleus. Assuming that the charge is uniformly distributed in the distorted nucleus, it can be shown that the barrier at the neck of the nucleus is reduced only in certain types of distortions. The barrier at the neck is always lower than other parts of the nucleus, if it is assumed that the charges in the nucleus concentrate themselves towards the ends of the deformed nucleus. The polarization of charge can be pictured in the following manner. As distortion of the nucleus takes place, the protons in the system move to either end of the deformed nucleus with respect to the neutrons. At later stages of distortion the interior charge will have taken a figure-eight distortion and finally will form two concentric charge systems as can be expected in two fragments just before separation. If this picture of charge polarization is true it may partly explain why no protons are emitted from the neck. One can assume that the barrier penetration factor, approximated by $\sigma_c^{\text{def}}(E)$ for the case of a deformed nucleus, is more than that for the spherical nucleus by a certain amount which takes into account the deformation of the nucleus. This can be written in the form

$$\sigma_c^{\text{def}}(E) = \sigma_c^{\text{sph}}(E) + A\sigma_c^{\beta}(E), \quad (7)$$

where A is an arbitrary constant depending on the degree of deformation, the region over which the depression takes place, etc. For no deformation of the nucleus, $A=0$. $\sigma_c^{\beta}(E)$ is the inverse cross section for the case of an electrically polarized deformed nucleus in an extreme case where the total charge of the nucleus is simulated by two point charges separated by a distance 2β. In the calculation of $\sigma_c^{\beta}(E)$ the lower cutoff radius was determined by assuming a constant volume for spheroidal distortion. The curves of $\sigma_c^{\text{sph}}(E)$, $\sigma_c^{\beta}(E)$, and $\sigma_c^{\text{def}}(E)$ vs E are shown in Fig. 6 calculated for $r_0 = 1.2$ F, along with the curve of $\sigma_c^{\text{sph}}(E)$ vs E as calculated by Blatt and Weisskopf for $r_0 = 1.3$ F. High-energy electron scattering experiments of Hofstadter seem to indicate that the value of $r_0 = 1.2$ F is justified for heavier nuclei. It is expected that for alpha particles of energy higher than about 18 MeV, where the barrier

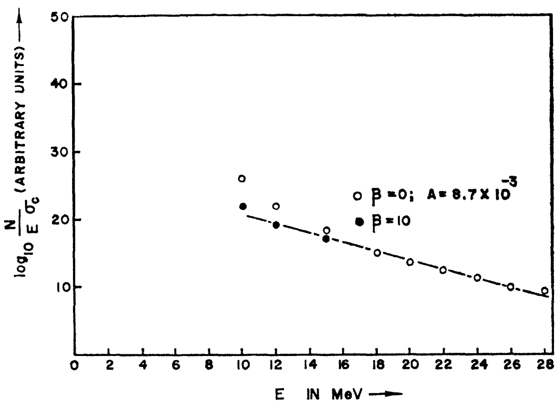

FIG. 7. The plot of $\log_{10}[N/E\sigma_c^{\text{def}}(E)]$ as a function of E.

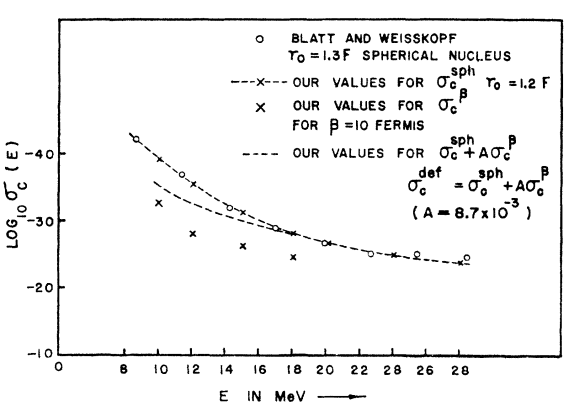

FIG. 6. The plot of $\sigma_c^{\text{sph}}(E)$ and $\sigma_c^{\beta}(E)$ as a function of E for different cases.

penetration factor nearly approaches unity, $\sigma_c^{\text{def}}(E)$ should be nearly the same as $\sigma_c^{\text{sph}}(E)$. This has been used to determine A. The $\sigma_c^{\beta}(E)$ was calculated for $\beta=10$. This value of β corresponds roughly to the diameters of the fragments. Figure 7 shows a plot of the experimental $\ln[N(E)/E\sigma_c^{\text{def}}(E)]$ against E, where $N(E)$ is the number taken from the measurements of Fulmer and Cohen.[7] Except for very low energies, the plot is linear. The dashed curve is a least-squares fit to the points and gives a distribution temperature, $T_D=0.63$ MeV.

The yield for the very low energies of the alpha particles (about 8 MeV) is undoubtedly higher than expected on the basis of our calculations on statistical evaporation of alpha particles. This discrepancy at low energies from the predictions of the evaporation theory has also been observed in other reactions[17–19] like (np), $(n\alpha)$, and (p,α) where one necessarily believes that the charged particles are evaporated. A general explanation for this anomaly is required for all such cases where charged particle emission takes place. This situation, therefore, does not act against the possibility of the alpha particles being evaporated.

Figures 3 and 4 show the angular distributions of fission fragments with respect to the incident neutron direction for binary and ternary fission. As expected from the theory of Bohr,[20] Halpern and Strutinski,[21] and others, the angular distribution for the binary fission is of the form $N(\theta)=1+\sum_n A_n P_n(\cos\theta)$. The fission fragment angular distribution in ternary fission is different from that of binary fission fragments. If the emission of alpha particles takes place after the nucleus has passed the saddle point, the distribution of $\bar{K}$ (the projection of $\bar{I}$ along the fragment separation direction) will remain unaffected, as the $\bar{K}$ distribution is assumed to be frozen in at the saddle point. One would have expected a slight reduction in the anisotropy due to the decrease and disorientation produced in $\bar{I}$ due to the emission of alpha particles. The emission of alphas before the saddle point would have resulted in somewhat higher anisotropy due to the decrease of K_0^2.

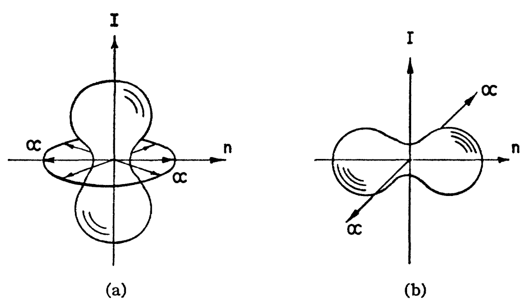

FIG. 8. The relative directions of emission of fission fragments and long-range alpha particles with respect to the incident neutron direction (a) when $K=K_{\max}$, (b) when $K=0$.

The experimental neutron fragment angular distribution in ternary fission results from the anisotropic $n-\alpha$ distribution and the fact that the alpha particles are favorably emitted from the neck, perpendicular to the line of flight of the fragments, where the barrier is somewhat depressed. This can be seen in the following way. The alpha particles are favorably emitted in a plane perpendicular to $\bar{I}$ [Figs. 8(a) and (b)] as indicated by the $n-\alpha$ angular distribution. For the cases of the fragment axis being perpendicular to $\bar{I}$ ($K=0$ and 0° emission), azimuthal directions of emission of alpha particles other than about perpendicular to neck are less probable due to high potential barrier in other directions. For the cases where the fragment axis is along $\bar{I}$ ($\bar{K}=\bar{K}_{\max}$ and 90° emission), all the azimuthal directions of emission of alpha particles in a plane perpendicular to $\bar{I}$ are favorable due to a lower potential barrier at the neck. This situation leads to a higher probability of alpha emission for fragments emitted at 90° as compared to those emitted at 0° with the incident neutron direction. The fact that directions of emission of alpha particles other than perpendicular to $\bar{I}$ are also probable, depending on the degree of anisotropy of the $n-\alpha$ distribution, will somewhat reduce this favorable emission of alphas from fragments emitted at 90° with the incident direction. If the inherent fragment distribution (as in binary fission) is forward peaked, the above situation will wipe out the anisotropy in the $n-f$ distribution for the cases of alpha emission. This has been found to be so experimentally (Figs. 3 and 4). This supports the hypothesis that the alpha emission takes place as an evaporation process from the neck of the distorted nucleus.

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