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Light-charged-particle emission in the spontaneous fission of ^{250}Cf, ^{256}Fm, and ^{257}Fm

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We have measured the energy spectra for the emission of long-range α particles from the spontaneous fission of ^{250}Cf, ^{256}Fm, and ^{257}Fm, and for tritons and protons from the spontaneous fission of ^{250}Cf and ^{256}Fm. We have determined α, triton, and proton emission probabilities and estimated total light-particle emission probabilities for these nuclides. We compare these and known emission probabilities for five other spontaneously fissioning nuclides with the deformation energy available at scission and show that there is a possible correlation that is consistent with a one-body dissipation mechanism for transferring release energy to particle clusters.

INTRODUCTION

Two suggestions for a mechanism for the formation and escape of a light-charged particle (LCP) during scission have been proposed by Halpern[1] and Carjan.[2] Halpern postulates that a sudden collapse of the newly distorted fragments to more spherical shapes might enable individual particles to acquire sufficient energy from the rapidly changing nuclear potentials in the neck region to become unbound. Carjan suggests that a preformed particle cluster could, through the one-body dissipation mechanism,[3,4] acquire sufficient energy to be emitted during the descent of the fissioning nucleus toward the scission point. Both of these hypotheses require that the energy supplied to emit the LCP be stored in the potential energy of deformation, either in a nascent fragment as the neck stub rebounds following rupture,[1] or in the fissioning nucleus as it stretches toward scission.[2]

We undertook the measurement of the LCP emission probabilities for the spontaneous fission (SF) of ^{250}Cf, ^{256}Fm, and ^{257}Fm for two reasons: We wanted to extend the range over which this probability is known to determine more reliably if there is a correlation with deformation energy, and we wanted to see if there was a noticeable reduction in the emission probability for ^{256}Fm and ^{257}Fm because of the onset of mass-symmetric SF in these nuclides, which would imply more spherical fragments and less deformation energy.

In 1947, Alvarez[5] reported on earlier work on the emission of long-range charged particles accompanying the fission induced in a foil of ^{235}U by slow neutrons. Since then, light-charged-particle emission during fission has been characterized to varying degrees for fission resulting from the reactions $^{233}\mathrm{U}+\mathrm{n}$ (Refs. 6–8), $^{235}\mathrm{U}+\mathrm{n}$ (Refs. 6–19), $^{235}\mathrm{U}+\gamma$ (Ref. 20), $^{238}\mathrm{U}+\mathrm{p}$ (Ref. 21), $^{239}\mathrm{Pu}+\mathrm{n}$ (Refs. 7, 22 and 23), $^{241}\mathrm{Pu}+\mathrm{n}$ (Ref. 6), $^{242}\mathrm{Am}^{m}+\mathrm{n}$ (Ref. 23), and the SF of ^{240}Pu (Ref. 6), ^{242}Pu (Ref. 6), ^{242}Cm (Refs. 6 and 24), ^{244}Cm (Ref. 6), and ^{252}Cf (Refs. 25–38). This list of references is by no means comprehensive, but it is representative of the work that has been performed on LCP fission.

From these studies, properties for LCP emission emerge that are remarkably invariant over the considerable range of fissioning nuclides investigated. The shape of the energy distribution for a given LCP remains essentially constant over this range, as does the angular distribution. This is somewhat surprising, because both the final energy and the angle of emission of the LCP result from the Coulomb repulsion of the LCP by the fission fragments, whose mass and charge vary considerably in going from the fission of the lighter actinides to that of the heavier actinides. The only LCP emission parameter that seems to vary with any statistical significance over the range of fissioning nuclides studied is the total LCP emission probability (or, as it has been called, the ratio of ternary to binary fission). The emission probability has been correlated with Z^2/A (the ratio of the electrostatic energy and the surface energy for a charged liquid drop) by Nobles,[6] and with the "removal energy" by Halpern[1,39,40] and Whetstone and Thomas;[25] the removal energy is a combination of the binding energy of the LCP to a fission-fragment nucleus and the reduction in the Coulomb potential of the fragments by the LCP between them. Under assumptions about the fragment emitting the LCP and the fragment separation distance at scission, the removal energy amounts to at least 20 MeV for all LCP's. However, the LCP emission probability does not seem to be uniquely dependent on removal energy for even a single fissioning nuclide. Halpern has also noted an empirical dependence of the LCP emission probability on the parameter $4Z-A$, with a family of straight lines depending upon the excitation energy of the fissioning nucleus.

Angular distributions measured for LCP emission (Refs. 14, 16, 17, 21, 27, 29, 32, 34, and 37) show that, in the laboratory frame of reference, LCP's are emitted at close to right angles to the fission-fragment axis. This result indicates that they must be emitted before the nascent fragments have attained any significant fraction of their final velocities; otherwise, the emission direction would be collinear with the fragment axis, as is the case for prompt fission neutrons. This implies that emission takes place either at or before scission, and, therefore, any mechanism that supplies the LCP with sufficient energy to surmount the nuclear and Coulomb barriers requires a readily available conversion of the potential energy of the fissioning nucleus as it descends from saddle to scission.

The energy acquired by the fissioning nucleus as it descends from its ground state toward the scission point is distributed among several degrees of freedom: internal heating, consisting of nontranslational collective (vibrational, rotational) and single-particle motions; prescission kinetic energy, from the translational motion of the two lobes of the nucleus as it stretches and the neck forms; and deformation potential energy, involving the change in shape of the nucleus as it becomes distorted. Deformation potential energy at scission is divided unevenly between the two fragments and is dissipated after scission into internal heating to be released as neutron and gamma-ray emission. Prescission kinetic energy is included in the measured kinetic energy of the fragments and has not been explicitly determined experimentally. Schultheis and Schultheis[41] have calculated that the internal heating at the scission point for the SF of ^{252}Cf is not likely to be more than about 30% (and could be considerably less) of this energy acquired during the descent toward scission. This essentially rules out the mechanism of particle evaporation by a hot nucleus, because the fission fragments at or before scission are rather cold and do not possess enough internal energy to carry out this process. Another observation that speaks against the evaporation mechanism is the negative dependence of the LCP emission probability on the excitation energy of the fissioning nucleus. Because the excitation energy contributed by the bombardment of a target nucleus with a projectile is in the form of internal excitation energy,[42] the emission probability should be strongly dependent upon this energy if the LCP is evaporated. In fact, the bombardment of lighter actinides by neutrons with energies between thermal and 14 MeV actually results in a slight decrease in the emission probability as compared with that for SF.[6,43,44]

EXPERIMENT

We constructed a light-particle counting system for this experiment consisting of a vacuum chamber in which two $\Delta E - E$ counter telescopes were mounted, one on either side of the sample. The ΔE detectors were fully-depleted silicon surface-barrier transmission detectors with nominal thicknesses of 86—90 μm; the E detectors were depleted to 1000 μm. All ΔE signals between 0.5 and 15 MeV and all E signals between 0.5 and 30 MeV arriving within the coincidence resolving time of 550 ns were accepted for each counter telescope independently, digitized, and stored on disks for subsequent off-line analysis.

All of the counting samples were prepared by evaporation of a chemically purified solution of the isotope onto 0.1 and 0.2 mg/cm^2 carbon foils attached to thin stainless-steel disks. Care was taken to obtain massless sources of a well-defined, reproducible geometry. The ^{256}Fm was produced via the (α,2n) reaction on ^{254}Es^g at the 88-inch cyclotron at the Lawrence Berkeley Laboratory; the ^{256}Md and its EC-decay daughter ^{256}Fm were then chemically separated from the other products of the reaction. The ^{250}Cf was obtained in a radiochemically pure form by milking it from its ^{254}Es^g grandparent; ^{254}Es^g, ^{257}Fm, and the ^{252}Cf employed as a comparison standard were used directly after chemical purification. Because of the very small SF branching ratios of ^{250}Cf (0.077±0.003%, Ref. 45) and ^{257}Fm (0.210±0.004%, Ref. 46), the decay-α particles from these isotopes can cause interfering reactions such as (α,p) with low-Z materials in the vicinity of the source, most notably the Si in our ΔE detectors. These light particles can contribute a significant background to the total detection signal. To reduce this effect, we degraded the energy of the decay α's below the Coulomb barrier for silicon + α by covering both sides of these sources with 2—3 mg/cm^2 carbon foils. No degrader foils were required for the ^{256}Fm source, which has a 91.9% SF branch.[45] We measured the SF activity of the ^{252}Cf standard and the ^{250}Cf and ^{257}Fm sources by counting them in an ion chamber and calculated the number of SF decays that had occurred during the LCP counting period from this activity. The number of SF decays occurring during the counting of the ^{256}Fm source could not be obtained this way, however, because of the rapidly varying composition of the sample. Therefore, we measured the efficiency of the ΔE detector facing the sample spot on the carbon mounting foil with an α standard of known activity and a spot size similar to that of the ^{256}Fm source. We counted the fission events from ^{256}Fm above 15 MeV in this detector with a scaler.

We calibrated our ΔE and E detectors with a precision pulse generator and an α-energy standard containing ^{148}Gd ($E=3.183$ MeV) and ^{228}Th and daughters ($E=5.423$, 5.686, 6.288, 6.779, and 8.784 MeV). The calibrations were checked frequently with the pulser and proved to be quite stable over a year of counting. We determined counting efficiencies for our two counter telescopes of 5.9±0.1% and 8.2±0.1% by removing the ΔE detector from each telescope and counting an α source of known activity with the E detectors in the singles mode. One of the counter telescopes required a collimator in the ΔE detector position, because the outer edge of this detector limited the acceptance angle of the E detector.

EXPERIMENTAL RESULTS

All LCP raw counting data were reduced by computer to give the energy deposited in the ΔE and E detectors for a coincident event; these energies were corrected for absorption in the degrader foils if they were used, employing the range-energy tables of Williamson *et al.*[47] In Fig. 1, we show the summed long-range α (LRA) energy distributions for three ^{252}Cf standard counts with degrader foils (a), and for two counts with no degrader foils (b). The LRA energies agree well with each other and are in agreement with published values for the peak LRA energy.[25–27,32,35,36] The energy distribution for the counts with the degrader foils [Fig. 1(a)] is slightly broader, as would be expected because of increased scatter in the foils. Because of the lack of collimation of our counter telescopes and the thickness of our ΔE detectors, we did not accept events from LRA's with total energies less than 13 MeV with no degrader foils and 14 MeV with degrader foils.

An example of our raw LCP counting data for ^{250}Cf is shown in Fig. 2 in a plot of ΔE energy versus total particle energy. The envelopes plotted are the limits within

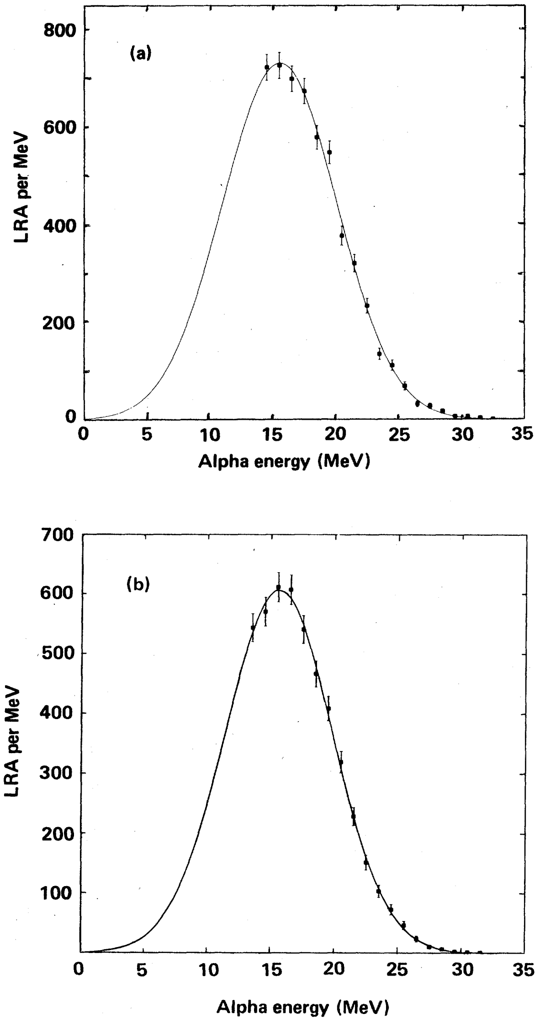

FIG. 1. Energy distributions for LRA from ^{252}Cf SF taken (a) with degrader foils, and (b) with no degrader foils.

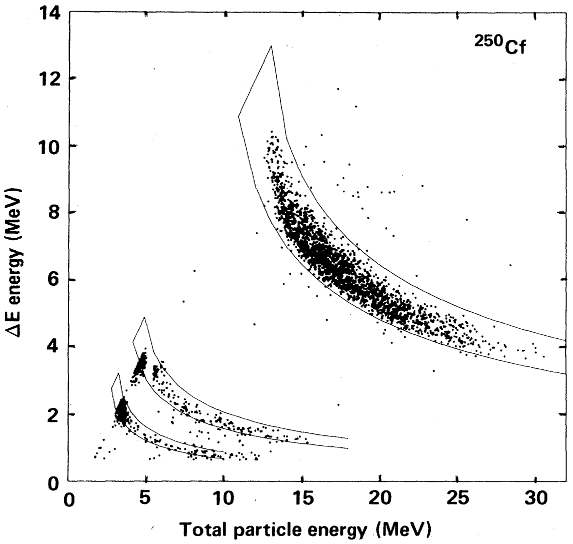

FIG. 2. Light-charged-particle data from this experiment for ^{250}Cf SF. The envelopes plotted are the limits within which the alphas, tritons, and protons should fall based on known range-energy relationships for these particles in Si and the geometry of the counter telescopes.

which the α's, tritons, and protons should fall based on the known range-energy relationships of these particles in Si and the geometry of our counter telescopes. Table I shows our results for the LCP energy spectra we obtained and the emission probabilities we calculated based on our counting geometry and SF counting rates.

Figure 3, a plot similar to Fig. 2, shows our counting data for ^{257}Fm. The numerous events in the energy region below that characteristic of the LRA's seem to be scattered somewhat randomly throughout that region; this, unfortunately, renders the triton and proton data for

TABLE I. Results of counting experiments for ^{250}Cf, ^{252}Cf, ^{256}Fm, and ^{257}Fm.

Nuclide	LCP (number observed)	E_{avg} (MeV)	FWHM (MeV)	LCP/10^3 SF
^{252}Cf (std)	alpha (9471)	15.6±0.2	10.3±0.5	3.21±0.46
	triton (860)	7.7±0.4	8.2±0.9	0.25±0.05
	proton (176)	7.9±0.4	6.7±2.3	0.05±0.01
^{250}Cf	alpha (4023)	16.1±0.2	10.0±0.9	3.98±0.28
	triton (273)	6.9±0.4	10.2±1.1	0.27±0.05
	proton (116)	8.2±0.2	6.6±1.4	0.09±0.02
^{256}Fm[a]	alpha (804)	15.5±0.4	11.3±1.0	4.62±0.59
	triton (66)	6.1±0.7	10.1±2.6	0.39±0.05
	proton (13)	6.6[a]	7.0[a]	0.07±0.02
^{257}Fm[b]	alpha (1169)	15.9±0.6	10.2±0.7	3.76±0.30

[a] Because of the low number of proton events, the energy distribution parameters from Ref. 36 were used in determining the emission probability for ^{256}Fm long-range protons.

[b] Triton and proton distributions were obscured by background events (see the text).

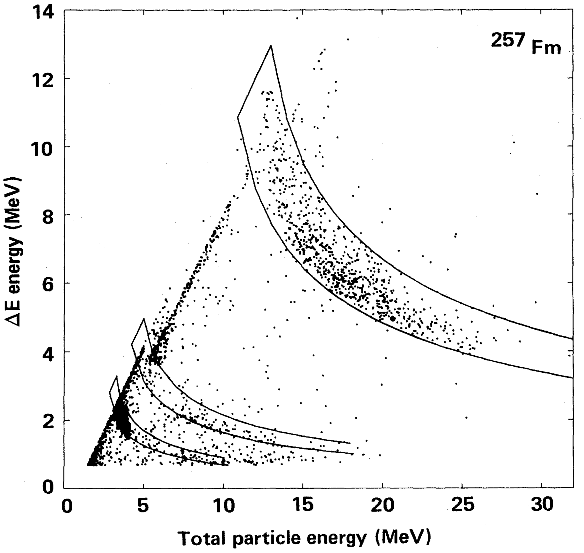

FIG. 3. Same as Fig. 2 but for ^{257}Fm SF.

^{257}Fm unusable. We cannot offer an explanation for this problem. The carbon degrader foils reduced the energy of the decay α's (6.63 MeV) of the granddaughter ^{253}Es more than 1 MeV below the Coulomb barrier for α's on silicon, which is 6.6 MeV. If the phenomenon were count-rate dependent, we would most likely have observed it also in the ^{250}Cf data, because the ^{250}Cf source strength was over 10^6 α/min, while the maximum count rate for the ^{257}Fm source was only 16 000 α/min, including daughters. If the events were from tritons or protons, their energies should have fallen principally within the range-energy envelopes for these particles. In determining the LRA emission probabilities in Table I, we corrected for the LRA emission below our cutoff energy using the data of Loveland,[35] who measured the LRA energy distribution for ^{252}Cf down to 0.5 MeV. Loveland demonstrated that the shape of this distribution is not entirely Gaussian, but is enhanced in the energy region below about 12.5 MeV. We fit his data to a composite of three Gaussian curves and determined analytically the fraction of LRA's emitted below a certain energy. We assumed that the energy distributions for tritons and protons were entirely Gaussian in shape and extrapolated those distributions below our cutoff energy, which was 6 MeV for both particles for the SF of ^{250}Cf and ^{252}Cf and 5 MeV for both particles for the SF of ^{256}Fm. Our results for the LRA, triton, and proton emission probabilities for ^{252}Cf agree reasonably well with published values.

Figure 4 shows plots of the energy distributions for LRA from ^{250}Cf, ^{256}Fm, and ^{257}Fm. Because we did not measure the emission probabilities for all LCP's from these isotopes, we assumed that the ratio of the LCP emission probabilities we did measure for each isotope is in the same ratio to the total LCP emission probability as it is for ^{252}Cf. Using this ratio, we converted our measured probabilities to total LCP emission probabilities for each of the isotopes we studied. We based this ratio on the in-

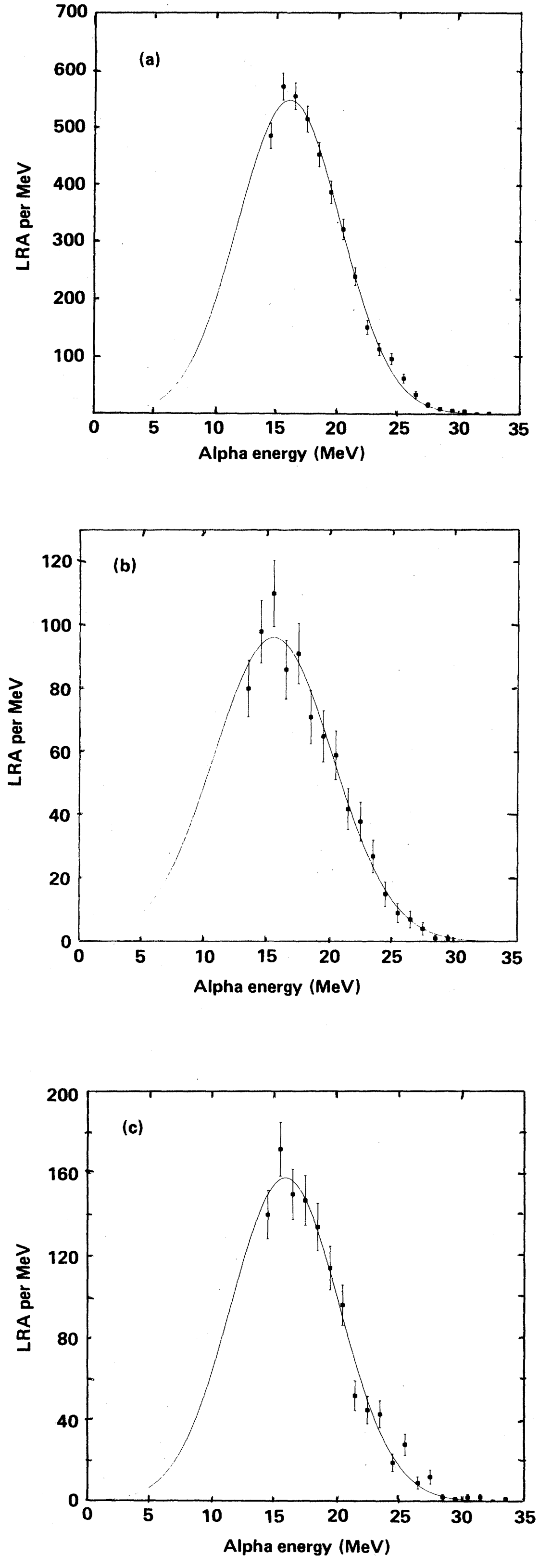

FIG. 4. Energy distributions of LRA measured in this experiment for the SF of (a) ^{250}Cf, (b) ^{256}Fm, and (c) ^{257}Fm.

TABLE II. Light-particle emission probabilities for ^{252}Cf SF[a].

LCP	Emission probability (per 10^3 SF)
alpha	3.334 ±0.11
triton	0.243 ±0.017
deuteron	0.022 ±0.002
proton	0.062 ±0.003
^{6}He	0.086 ±0.018
^{8}He	0.0031±0.0003
Li	0.0039±0.0002
Be	0.0129±0.0048
	Sum = 3.767 ±0.113

[a]Weighted average of values from Refs. 26 and 29, except for the alpha (from Ref. 29 only) and ^{6}He (weighted average of Refs. 26 and 36).

formation shown in Table II, which is a composite of the most thoroughly determined ^{252}Cf emission probabilities found in the literature. It can be seen that the ratio of LRA to LRA + tritons + protons for ^{252}Cf from Table II is 0.916±0.005; the same ratios for our measurements given in Table I are 0.915±0.017, 0.917±0.013, and 0.909±0.014 for ^{252}Cf, ^{250}Cf, and ^{256}Fm, respectively, in excellent agreement. The ratio of LRA + tritons + protons to total LCP's in Table II for ^{252}Cf is 0.966±0.005; we used this ratio and our measured LCP emission probabilities to obtain values of total LCP/10^3 SF of 4.49±0.30 and 5.26±0.61 for ^{250}Cf and ^{256}Fm, respectively. To obtain the total LCP emission probability for ^{257}Fm, for which we measured only LRA, we used the value of LRA/LCP from Table II of 0.885±0.007 to calculate a value of 4.25±0.34 LCP/10^3 SF for ^{257}Fm.

DISCUSSION

Halpern's theory[1] of LCP emission, in which energy for releasing a LCP is acquired from the rebound of the neck stub into a fragment after scission, was presented in a qualitative sense; however, estimates suggest that the time required to collapse the neck stubs is not likely to be short enough to transfer enough energy to the LCP before the fission fragments are considerably accelerated.[42] Carjan[2] has quantified somewhat his theory for LRA emission, which postulates that heavy nuclides (including, of course, those for which LRA emission has been observed) are α emitters not only in their ground states, but also all along the way to the scission point. They are, therefore, capable of preforming α clusters at any instance between the saddle and scission points. His mechanism involves the collision of these preformed α clusters with the inside of the nuclear surface (α-nucleus potential) in the region of the neck (one-body dissipation[3,4]). As the cluster rebounds from the "wall," if it does not dissolve into its constituent nucleons, it then moves toward the opposite wall, collides, and rebounds again, gaining more energy. Clusters that are formed late in the fission process in the neck region can acquire sufficient energy to surmount the potential barriers (nuclear and Coulomb) and escape. The amount of energy gained with each collision of the cluster and the wall is a function of the velocity of the nuclear surface, which is in turn a function of the deformation energy and the rate of distortion. Thus, it is reasonable to assume that those fissioning systems that possess larger amounts of deformation energy will have higher LRA emission probabilities. Because these heavy nuclides have an almost spherical saddle point shape, the more stretched the scission point configuration is, the longer it takes to reach the scission point, and the more opportunities there are to emit a LRA during the transition from saddle to scission. The energy transferred to the α cluster comes at the expense of prescission fragment kinetic energy. Prescission kinetic energy, as stated before, is included along with the fragment kinetic energy from Coulomb repulsion as the total kinetic energy (TKE). Therefore, the TKE for fission accompanied by LCP emission should be less than that for binary fission, which is the case as measured for ^{235}U(n,*f*) (Ref. 11) and ^{252}Cf SF,[33] with reductions in the postneutron average TKE of 13.4 and 13.6 MeV, respectively. This is more than can be accounted for by the decrease in Coulomb repulsion from just the loss of two protons and two neutrons from the fissioning system. Carjan's theory also applies to the other light charged particles, although their existence as free entities in the neck region of the fissioning nucleus is less likely and may be one reason why their emission probabilities are lower.

A good estimate of the deformation potential energy at scission can be made by subtracting the measured average TKE from the calculated fission Q value. This difference should equal the deformation energy plus the internal heating, which we assume is low in SF from the theoretical estimates.[41] We list in Table III the LCP emission probabilities we measured along with those from five other SF nuclides that have previously been measured, and the corresponding values for $\langle Q\text{-TKE}\rangle$ for each nuclide. The Q values were calculated from the Comay-Kelson mass excess values[48] averaged over the experimental mass distribution. Fragment atomic numbers were calculated using the prescription of Nethaway.[49] Throughout this article, literature values of TKE, including those in Table III, measured based on ^{252}Cf calibrations employing the parameters of Schmitt, Kiker, and Williams[50] were reduced by a factor of 1.0104 to conform to the redetermination of these parameters by Henschel *et al.*[51]

We have listed only SF-emitting nuclides to avoid any possible effects from excitation energy contributed by bombarding projectiles used to induce fission. The LCP emission probability for ^{242}Cm is a weighted average value; the ^{242}Cm results of Perfilov *et al.*[24] were obtained using nuclear emulsions to record the light particles. An absorbing foil between the SF source and the emulsion resulted in a cutoff α energy of 11 MeV. We corrected for the unobserved portion of their α spectrum and for the likelihood that they also observed the other LCP's above the cutoff energy characteristic of each type of particle. With these corrections, the values of Nobles[6] and Perfilov *et al.* for ^{242}Cm agreed quite closely. The residual energy values of ^{240}Pu (Ref. 59) and ^{252}Cf (Ref. 60), for which the total fission energy balance has been measured experimentally, are in good agreement with the $\langle Q\text{-TKE}\rangle$ values in

TABLE III. LCP emission probabilities and estimated deformation potential energy for SF-emitting nuclides.

Nuclide	LCP/10^3 SF	Q (MeV)	Avg TKE (MeV)[a,b]	$\langle Q\text{-TKE}\rangle$ (MeV)
^{240}Pu	3.18±0.20[c]	199.7	177.2±0.5[d,e]	22.5±0.5
^{242}Pu	2.74±0.22[c]	200.5	179.9±0.5[d,f]	20.6±0.5
^{242}Cm	3.91±0.23[c,g]	210.8	181.1±2.4[h,i,j]	29.7±2.4
^{244}Cm	3.18±0.20[c]	210.1	181.8±2.0[k]	28.3±2.0
^{250}Cf	4.49±0.30[l]	220.5	185.1±0.5[i]	35.4±0.5
^{252}Cf	3.77±0.11[m]	219.1	184.1±1.3[n]	35.0±1.3
^{256}Fm	5.26±0.61[l]	234.7	196.9±0.5[o]	37.8±0.5
^{257}Fm	4.25±0.34[l]	236.3	197.1±0.5[p,q,r]	39.2±0.5

[a] Average preneutron TKE.

[b] References are for average TKE values only.

[c] Reference 6.

[d] Error on average TKE increased to 0.5 MeV for ^{240}Pu and ^{242}Pu.

[e] Reference 52.

[f] Reference 53.

[g] Reference 24.

[h] Average between $Z^2/A^{1/3}$ systematics of Unik *et al.* (Ref. 54) and Viola (Ref. 55).

[i] Reference 54.

[j] Reference 55.

[k] Reference 56.

[l] This work.

[m] See Table II.

[n] Reference 51.

[o] J. F. Wild and E. K. Hulet, unpublished data, Lawrence Livermore National Laboratory, 1984.

[p] Data from Ref. 57 reanalyzed with a different neutron-emission correction obtained from the data of Ref. 58.

[q] Reference 57.

[r] Reference 58.

Table III based on the old Schmitt-Kiker-Williams calibration.[50]

These data are presented in Fig. 5 as a plot of LCP emission probability *vs* $\langle Q\text{-TKE}\rangle$ (as an estimate of deformation energy). Although there is a considerable amount of dispersion in the measured LCP emission probabilities about the linear least-squares fit, there is reason to suggest a direct correlation between the emission probability and the deformation energy at scission. A positive experiment to demonstrate the validity of this hypothesis would be the measurement of the LCP emission probabilities for the SF of ^{258}Fm and ^{259}Fm. The SF of ^{258}Fm (Ref. 61) and ^{259}Fm (Ref. 62) exhibits average fragment TKE's (235 and 240 MeV, respectively) that are uniquely higher by 40 MeV than any others yet measured. These TKE values, which are near the Q value for the fission process, imply that there can be only very little deformation energy available, and the fission fragments must be nearly spherical. Thus, there is little energy available for LCP formation and escape, and the emission probabilities must be quite low. Unfortunately, this measurement would be a difficult undertaking, because of the short half-lives of these isotopes and the problem of producing sufficient amounts to make reliable measurements. It might be possible to make a sufficient amount of ^{258}Fm for this measurement via production of its electron-capture decay parent, the 60-min isomer of ^{258}Md.

In sum, we have measured LCP emission probabilities and energy distributions for the SF of ^{250}Cf, ^{256}Fm, and ^{257}Fm. We find that there is a correlation between the emission probabilities and the available deformation energy during fission. We believe that this correlation is plausible and that Carjan's theory suggests a reasonable mechanism for LCP emission in fission.

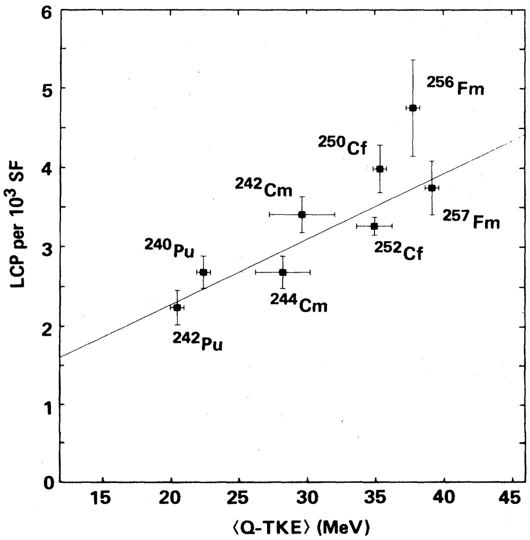

FIG. 5. Light-charged-particle emission probabilities from SF *vs* the deformation plus internal excitation energy at scission.

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