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Light Charged Particles from Spontaneous Fission of Cf^{252}[†]

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A semiconductor ΔE-E counter telescope was used to identify light charged particles spontaneously emitted from Cf^{252}. From the measured partial-energy spectra of the particles, it was possible to estimate by extrapolation the following total yields of the various particle types observed. These are (in number per fission): $H^1(>1.6\times10^{-4})$, $(H^1)(5.1\pm0.5\times10^{-5})$, $H^2(2.0\pm0.1\times10^{-5})$, $H^3(1.90\pm0.06\times10^{-4})$, $He^3(\lesssim2.9\times10^{-5})$, $He^4(3.27\pm0.10\times10^{-3})$, $He^6(7.8\pm1.6\times10^{-5})$, $He^8(5.9\pm1.6\times10^{-6})$, $He^{10}(3\pm3\times10^{-7})$, $Li(3.9\pm2.0\times10^{-6})$, $Be(>3\times10^{-7})$. The H^1 energy spectrum is qualitatively different from the spectra of the other particles in that it contains a large component that appears to increase greatly with decrease in energy at the lowest energies. The smaller component that remains after subtraction of this apparent background is denoted (H^1), and it is speculated that these are the "scission" protons. The yields were all obtained relative to the He^4 yield, the value for which was taken from Thomas and Whetstone. The energy spectra of (H^1), H^2, H^3, He^3, He^4, He^6, and He^8 were found to have maxima at 9 ± 2, 7 ± 2, 8 ± 1, 17 ± 1, 16 ± 0.5, 13 ± 1, and $\lesssim13$ MeV and full widths at half-maximum of 6 ± 2, 7 ± 1, 6 ± 1, 9.5 ± 0.5, 11.5 ± 0.5, 8 ± 1, and 8 ± 4, respectively. The yields correlate more or less as expected with Halpern's estimates of the energy required for release of the particles into the region between the fission fragments, and the dependence of the energy spectra on mass can be understood in terms of the Halpern model.

1. INTRODUCTION

THE relatively rare modes of fission in which a charged particle of low mass is emitted are of particular interest both to the theory of the fission process and to the theory of nuclear matter in general. This interest arises because studies of the angular distribution of alpha particles from fission indicate that these particles are released in the space between the two separating fragments very nearly at the time of the actual scission of the fissioning nucleus.[1–3] The observed light charged particles, in their probability of emission and in their energy and angular distributions, are thus expected to carry information imparted to them at their birth concerning the configuration of the nucleus at the time of scission, in particular, revealing the properties of the neck that presumably connects the incipient fragments just before scission.

The literature on the emission of light charged particles in fission is large, and only the briefest summary can be made here.[4] The first notice of the process and most of the earliest work was accomplished with nuclear emulsions, an experimental technique that is handicapped severely by backgrounds caused by nuclei of the constituents of the emulsion recoiling from collisions with the heavy, energetic fission fragments. Moreover, the relative rarity of the events of interest makes data accumulation by usual scanning methods slow. The greatest disadvantage, from the point of view of the present studies, is the lack of precision in the determination of the particle mass. It soon became apparent, however, that most of the long-range particles were alpha particles. Extensive measurements, many utilizing ionization chambers, have been reported on the emission probability, energy spectrum, and angular distribution of the alpha particles from a variety of fissioning systems.

Protons were reported as produced in the thermal-neutron-induced fission of U^{235} by Hill[5] in 1952. Fulmer and Cohen[6] detected tritons from the same system, but attributed them to reactions between lithium impurities and the fission neutrons. Tritium was identified radiochemically from uranium fission by Albenesius and Ondrejcin[7,8] and by Sloth, Horrocks, Boyce, and Studier[9] and from the spontaneous fission of californium by Horrocks.[10] Watson[11] and Wegner[12] used the $\Delta E\times E$ technique to identify tritons from the spontaneous fission of Cf^{252}; Wegner[12] also reported protons and upper limits for deuterons and He^3. He^6 and evidence for He^8, lithium, and beryllium from Cf^{252} were reported

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[†] Work performed under the auspices of the U. S. Atomic Energy Commission.

[*] Alfred P. Sloan Research Fellow.

[1] Tsien San-Tsiang, J. Phys. Radium **9**, 6 (1948).

[2] I. Halpern, "Alpha Particle Emission in Fission," CERN, Geneva (unpublished).

[3] I. Halpern, *Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965), Vol. II, p. 369.

[4] For reviews of the subject, often called "ternary" fission, see E. K. Hyde, *The Nuclear Properties of the Heavy Elements* (Prentice-Hall Publishers, Inc., Englewood Cliffs, New Jersey, 1964), Vol. III, pp. 131–140; W. J. Whitehouse, Progr. Nucl. Phys. **2**, 158 (1952).

[5] D. L. Hill, Phys. Rev. **87**, 1049 (1952).

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

[7] E. L. Albenesius, Phys. Rev. Letters **3**, 274 (1959).

[8] E. L. Albenesius and R. S. Ondrejcin, Nucleonics **18**, 100 (1960).

[9] E. N. Sloth, D. L. Horrocks, E. J. Boyce, and M. H. Studier, J. Inorg. Nucl. Chem. **24**, 337 (1962).

[10] D. L. Horrocks, Phys. Rev. **134**, B1219 (1964).

[11] J. C. Watson, Phys. Rev. **121**, 230 (1961).

[12] H. E. Wegner, Bull. Am. Phys. Soc. **6**, 307 (1961).

by Whetstone and Thomas.[13,14] The emission of He^8 from Cf^{252} has been confirmed by Cosper, Cerny, and Gatti[15] and recent measurements by Poskanzer, Esterlund, and McPherson[16] confirm the particle stability of He^8. Searches for particles of charge greater than 2 using radiochemical techniques or emulsions have yielded either negative results or very low upper limits for the production of these particles.

Halpern[2,3] has shown that a semiclassical model for third-particle formation is useful in accounting reasonably quantitatively for both the energy and angular distribution of alpha particles produced in fission. He has extended this model to give predictions of the amount of energy necessary for the production of other particles at scission. It is assumed that there is a correlation between these energies and the yields of the various particles; the higher the required energy, the lower the yield. Investigations of the angular and energy distributions and yields of these other particles provide tests for Halpern's model[2,3] and further restrictions on the possible scission configurations.

We report here on the yields of H^1, H^2, H^3, He^3, He^4, He^6, He^8, He^{10}, Li, and Be, all from the spontaneous fission of Cf^{252}. Sufficient data were obtained to provide energy distributions for all but He^{10} and Be. (We note that Garvey and Kelson,[17] using a mass formula that gives known masses to high accuracy, predict that He^{10} is unstable by 10 MeV with respect to two-neutron emission. It seems unlikely, therefore, that the two events we have seen are actually due to He^{10}.)

Neither this experiment nor any other known to us has established that the various light charged particles, other than He^4, observed from fissioning systems are actually emitted in coincidence with fission, or are emitted with an angular distribution with respect to the fission fragments like that which characterizes so strongly the alpha-particle emission. The energetic requirements for their production, however, are such as to lead to the conclusion that they must be associated with the fission process. Possible alternative mechanisms are secondary reactions induced by the fission neutrons, or, with lower probability, by the fission fragments or natural alpha particles incident on low Z nuclei, or evaporation from the excited fission fragments. The number of neutrons with energies high enough to produce 8-MeV protons or 12-MeV He^6 particles is, however, not large enough to account for the observed yields of these particles. Estimates by Grover and Thomas of the probability of evaporating protons from excited fragments in the thermal-neutron-induced fission of U^{235} give values that are considerably lower than the value observed for the spontaneous fission of Cf^{252}. Because the excitation energy of the fragments is higher for californium fission than for uranium fission, it is expected that the calculated yield of evaporated protons will also be higher. We cannot, at this time, exclude evaporation as a possible mechanism for the production of some of the particles.

2. EXPERIMENTAL METHOD

Two sources of Cf^{252} were deposited by evaporation from solution onto a platinum backing; one had an intensity of about 10^5 fissions/min, and the other about 10^6. To impede the self-transfer of californium to the vacuum chambers and detectors, the sources were covered with thin nickel foils. For the weaker source the thickness of this cover was 100 μg/cm^2, and for the stronger, 400 μg/cm^2.

The light charged particles were identified with a counter telescope consisting of two semiconductor detectors: a thin, totally depleted, "ΔE" detector followed by a thick "E" detector. An aperture in front of the ΔE detector ensured that any particle from the source that passed through the ΔE detector would, if unscattered, strike the E detector. The counter telescope system subtended an angle at the source of about 15° in some experiments and 7° in others. The ΔE detector was covered either with an aluminum foil or with a lead foil to stop fission fragments and particles from the alpha decay of californium from reaching the detector.

An aluminum foil thickness of 7.6 mg/cm^2 was found to be sufficient to stop essentially all of the fragments and 6.11-MeV alpha particles; foils 6.6 and 10.6 mg/cm^2 were also used in several runs to investigate background effects. A lead foil with a thickness of 30.8 mg/cm^2 was used in the studies of particles with $Z=1$ in order to reduce the probability of secondary reactions due to the abundant 6.11-MeV alpha particles. During the course of the measurements, ΔE detectors with thicknesses of 29, 42, 49, 62, and 112 μ, and E detectors, 400-, 500-, and 2000-μ thick,[18] were used. The thinner ΔE detectors permit particles of lower energy to be identified by mass, at a substantial sacrifice in mass resolution, however. The thinner E detectors used were thick enough to stop essentially all particles of charge $Z>1$ that could reasonably be expected to originate from spontaneous fission; the thicker detector stopped protons of energies up to 18 MeV and heavier particles to correspondingly higher energies.

The signals from the two detectors were converted to voltage pulses and amplified in conventional ways. The pulses corresponding to E and ΔE were summed to

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[13] S. L. Whetstone, Jr., and T. D. Thomas, Bull. Am. Phys. Soc. **10**, 722 (1965).

[14] S. L. Whetstone, Jr., and T. D. Thomas, Phys. Rev. Letters **15**, 298 (1965).

[15] S. W. Cosper, J. Cerny, and R. Gatti [unpublished work quoted by J. Cerny, S. W. Cosper, G. W. Butler, R. H. Pehl, F. S. Goulding, D. A. Landis, and C. Détraz, Phys. Rev. Letters **16**, 469 (1966)].

[16] A. M. Poskanzer, R. A. Esterlund, and R. McPherson, Phys. Rev. Letters **15**, 1030 (1965).

[17] G. T. Garvey and I. Kelson, Phys. Rev. Letters **16**, 197 (1966).

[18] The 29- and 62-μ thick detectors were actually detectors 28- and 60-μ thick, tipped 15° from the mean direction of particle incidence.

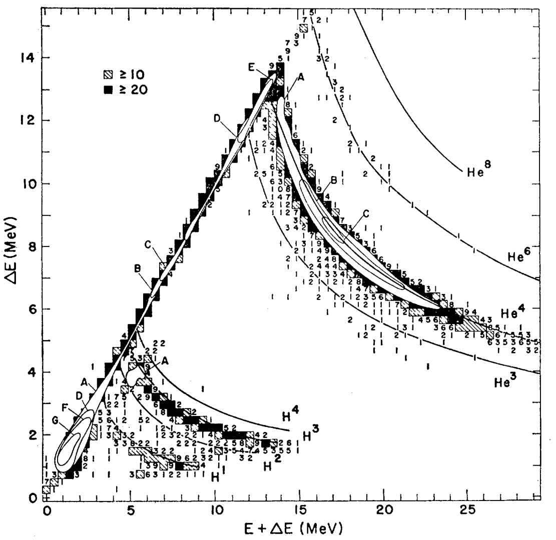

FIG. 1. $\Delta E\times(E+\Delta E)$ characteristics of particles emitted from Cf^{252}. Energies are as deposited in the detectors and do not include loss in absorber foil. Detector thicknesses: ΔE, 112 μ; E, 400 μ. The contour lines are drawn at 40 events (A), 80 (B), 100 (C), 140 (D), 200 (E), 1000 (F), and 2000 (G).

give a pulse whose height was proportional to the total energy of the detected particle. This pulse and that corresponding to ΔE were analyzed on the two axes of a two-parameter analyzer when they occurred in coincidence.

A typical display of the data is shown in Fig. 1, where the ordinate is proportional to the energy deposited in the ΔE detector and the abscissa to the total energy of the particle. The number of events recorded for each pair of pulse heights, E and $E+\Delta E$, is indicated. Particles that stop in the ΔE detector produce points that lie along the diagonal line corresponding to ΔE equal to $E+\Delta E$. Particles that penetrate into the E detector must deposit less than their full energy in the ΔE detector, and therefore produce points that lie below the diagonal line. The point of departure from the line and the locus of points for higher particle energy is dependent, via the range-energy relations, on the charge and mass of the particle and on the thickness of the ΔE detector.

We have used the easily identified locus for the He^4 particles and the range-energy tables of Williamson and Boujot[19] to determine the actual thickness of a given ΔE detector; from this thickness and the range-energy tables other loci are determined. Tables for He^6 and He^8 were constructed according to the adequate assumption that the ratio of the range to mass is represented by the same function of the velocity for all masses of particles with the same charge.

The energy calibration is based on the response of each of the detectors to the 6.11-MeV alpha particles from the alpha decay of Cf^{252} or to the 5.47-MeV alpha particles from Am^{241}, and on the response of the amplifier-analyzer systems to a pulser. Spectra of He^4 particles emitted in fission from the various runs were also intercompared to permit small adjustments in the calibration.

To investigate possible distortion of the data due to "channeling" effects[20] in the ΔE detector, we made a $\Delta E\times(E+\Delta E)$ measurement with a 60-μ thick ΔE detector tipped so that the normal to the center of its front surface made an angle of 15° with the line between the source and the axis of the counter telescope. A measurement was then made with the ΔE detector rotated 30° about the central normal to its surface. Since the detector was presumed to have been prepared (as was usual until recently) with its front surface normal to the (111) crystalline axis, this procedure should result in one of the two orientations being extremely unfavorable to the channeling of the incident particles through its crystal structure, despite our 7° half-angle of acceptance. No significant effects due to channeling were observed. The measurements of particles with $Z=1$ were made using a detector cut with an orientation chosen to minimize channeling effects.

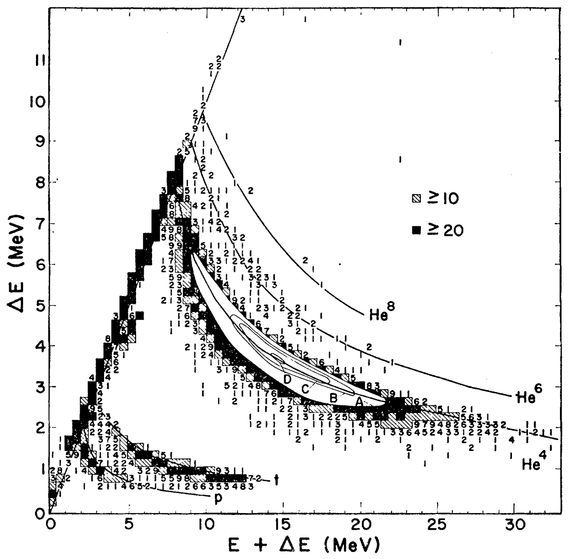

FIG. 2. $\Delta E\times(E+\Delta E)$ characteristics of particles emitted from Cf^{252}. Energies are as deposited in the detectors and do not include loss in absorber foil. Detector thicknesses: ΔE, 49 μ; E, 400 μ. The contour lines are drawn at 40 events (A), 80 (B), 100 (C), and 140 (D).

3. RESULTS

Examples of the $\Delta E\times(E+\Delta E)$ data are shown in Figs. 1–3 for ΔE detector thicknesses 112, 49, and 29 μ, respectively.[18] The energy scales, determined as described in Sec. 2, are believed to be accurate to a few

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[19] C. Williamson and J. P. Boujot, Centre d'Etudes Nucléaires de Saclay, Report No. CEA-2189, 1962 (unpublished).

[20] G. Dearnaley, IEEE Trans. Nucl. Sci. **NS-11**, No. 3, 249 (1964); C. Erginsoy, H. E. Wegner, and W. M. Gibson, Phys. Rev. Letters **13**, 530 (1964).

percent. Given in these figures are the energies deposited in the two silicon detectors; in each of the cases shown, the particles had to penetrate 7.6 mg/cm^2 of aluminum to reach the ΔE detector.

The greatest separation of the loci for the various particle types and the best energy resolution is obtained for the thickest of the detectors, but then, of course, more of the lower portion of the particle-energy spectra becomes lost as the particle identification fails. (The spectrum for He^4 particles is probably given with adequate accuracy down to much lower energies under the assumption that their yield predominates at low energies along the locus for $\Delta E = E + \Delta E$.)

It was not possible to extract useful information concerning each of the particle types from every run. For example, it is apparent in the case shown in Fig. 3 that only for particles of $Z \geq 2, A \geq 4$ is the identification sufficiently unambiguous. It is also evident that many of the $Z=1$ particles possess sufficient energy to penetrate the E detectors used in this experiment. This causes the locus of these particles on the $\Delta E \times (E+\Delta E)$ plot to fold back toward the origin, confusing the $E+\Delta E$ spectrum. Particles are also lost to the analysis if either their ΔE or $E+\Delta E$ pulse heights fall outside the bounds of the pulse-height analyzer. Some background is also certainly present due primarily to a small contamination of the ΔE detector by stray Cf^{252}, which gives predominantly 6.11-MeV alpha particles (usually observed as a peak in our data arrays) and also significant numbers of fission fragments and fast neutrons, which may cause secondary effects. Some of the background in the $Z=1$ region may in fact be attributed to such reactions as (n,p) and (n,α) on Al^{27} or Si^{28}.

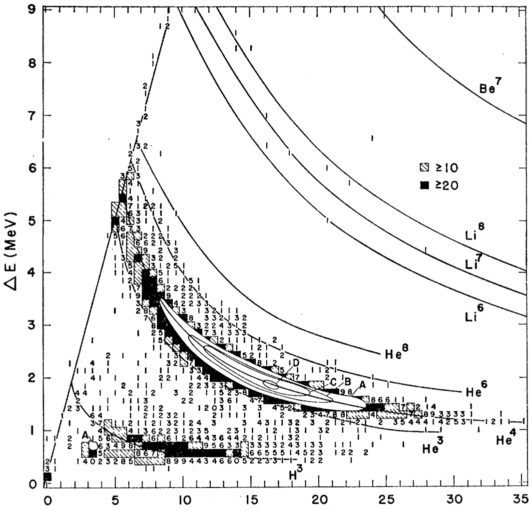

FIG. 3. $\Delta E \times (E+\Delta E)$ characteristics of particles emitted from Cf^{252}. Energies are as deposited in the detectors and do not include loss in absorber foil. Detector thicknesses: ΔE, 29 μ; E, 500 μ. The contour lines are drawn at 40 events (A), 80 (B), 100 (C), and 140 (D).

In Fig. 4 are shown data for the heavily ionizing particles of $Z \geq 2$ obtained with a 49-μ ΔE detector (behind 7.6 mg/cm^2 of aluminum) and lowered gain in the ΔE system. The loci drawn were in this case determined from the range-energy graphs of Northcliffe[21] for Li and Be nuclei in aluminum. Control runs (with the Cf^{252} source removed) totaling 38 h (compared with the 79 h of running time for the data shown in Fig. 4) yielded only one event (labeled "B.G." in Fig. 4) that did not lie on the $\Delta E = (E+\Delta E)$ line. It does not seem likely that the $Z>2$ events observed were caused by sporadic electronic misbehaviors, but rather that perhaps the one background event observed resulted from contamination or cosmic-ray effects.

The energy spectra for the identifiable particles are obtained by summing the appropriate portions of the arrays. Corrections are then made for the energy lost in the aluminum absorber. (The small amount of energy lost in the 100 μg/cm^2 nickel cover foil and in the small dead layers of the detectors is neglected.)

The data from 12 of the 20-odd runs, each providing information in intervals of energy (incident on the aluminum) of different position and width, and spanning different ranges, were combined by means of a coded calculation that performed a linear interpolation transforming to distributions with standard and uniform energy intervals. The data occurring in the standard intervals that overlapped the ends of the data range were either discarded or corrected. Although the statistical error to be assigned to the points is affected by the smoothing effect of the transformation, it was ignored. Finally, since the runs were of unequal length,

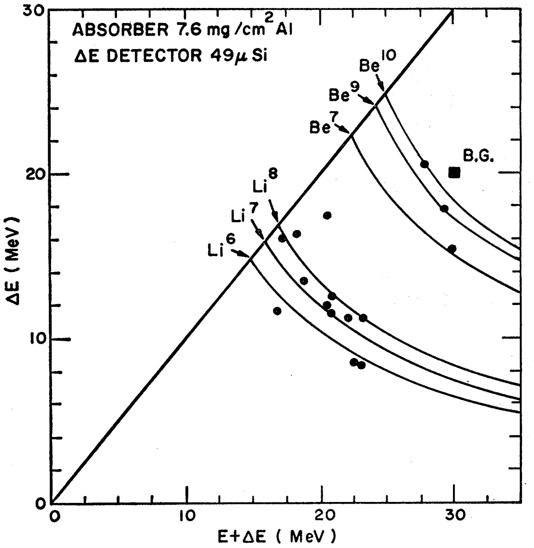

FIG. 4. $\Delta E \times (E+\Delta E)$ characteristics of *heavily ionizing* particles emitted from Cf^{252}. Energies are as deposited in the detectors and do not include loss in absorber foil. Detector thicknesses: ΔE, 49 μ; E, 400 μ.

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[21] L. C. Northcliffe, Nat. Acad. Sci.–Nat. Res. Council Nucl. Sci. Ser. Rept. No. 39, Publ. 1133 (1964), p. 180.

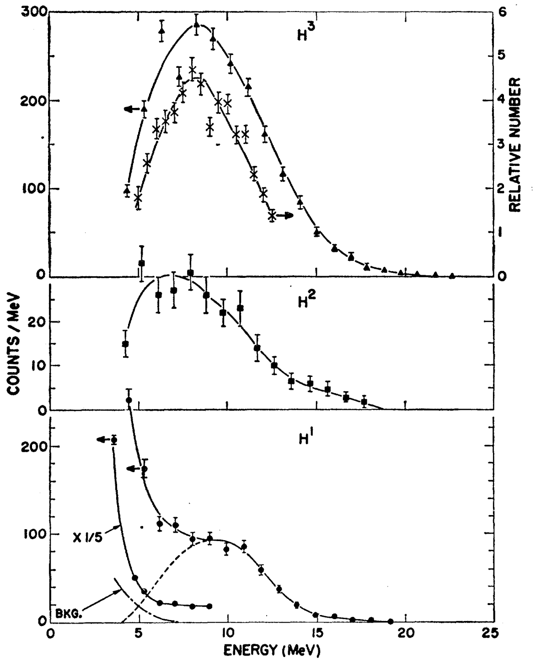

FIG. 5. Energy spectra for the particles with $Z=1$ and masses as noted corrected for loss in the aluminum absorber foil. The H^3 spectrum denoted by the crosses was measured with the smaller of the two Cf^{252} sources and with the thinner E detectors.

the individual distributions were normalized, before combination, to the total number of He^4 particles detected in the respective run. In all cases this entailed an extrapolation of the observed He^4 spectrum to zero energy using the spectral shape measured by Nobles.[22]

The spectra for the $Z=1$ particles H^1, H^2, and H^3 are shown in Fig. 5. An attempt was made to evaluate the proton background arising from secondary reactions of the fission neutrons by making measurements with a piece of $\frac{1}{32}$-in. stainless steel covering the source. The result is shown as the dot-dashed curve of Fig. 5, but must be viewed as a lower limit, since the steel absorber blocked some possible reaction sites from being seen by the detector. It appears that the observed H^1 spectrum may contain two components: a broad peak, much like that observed for the other particles and presumably associated with fission, superimposed on a large background that increases greatly with decreasing energy at the lowest energies. We have denoted by (H^1) in the tables and figures the component believed to be associated with fission. Spectra for $Z=2$ particles He^3, He^4, He^6, and He^8 are shown in Fig. 6. (Two possible He^{10} events were observed, one of them appearing in Fig. 3; however, see the comments in the Introduction concerning the likelihood that these are due to He^{10}.) The combined spectrum for Li^6, Li^7, and Li^8 obtained from the data arrays shown in Figs. 3 and 4 is given in Fig. 7.

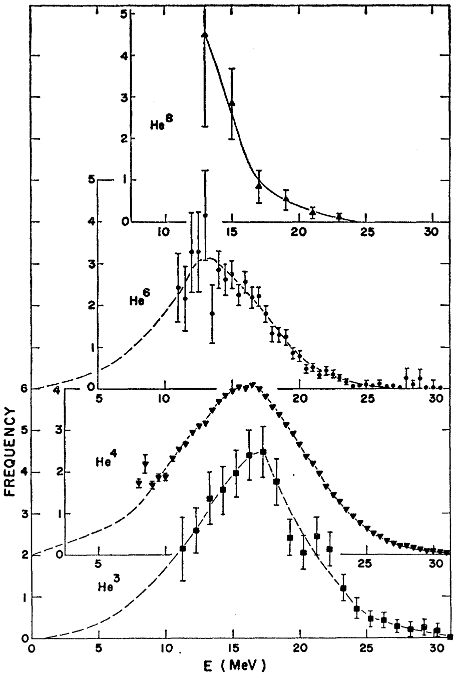

FIG. 6. Energy spectra for the particles with $Z=2$ and masses as noted corrected for loss in the aluminum absorber foil.

The information obtained on the total yields of each of the particle types is presented in Table I. For each particle type "x" the energy interval observed in the experiments, $\Delta E(x)$, and the number of identified particles used in the analysis, $n(x)$, are given. Next is listed the number of particles of type x identified in the given energy interval divided by the total number of He^4 particles of all energies emitted in fission (as determined from the extrapolations of the observed He^4 spectra). The uncertainties quoted are standard deviations based on the number of particles identified. Finally, estimates are given, where it seems possible, for the total number of particles of type x per He^4 emitted in fission, and, for convenience, the total number per fission.

4. DISCUSSION

Halpern,[2,3] in his analysis of the phenomenon of alpha-particle emission in fission (and its extension to the emission of other light charged particles by the same mechanism), has shown how the process can be considered to take place in two main, more or less independent, successive steps. The first involves the release of a charged particle into the space between the fission fragments; the second, the subsequent motion of the particle in the Coulomb field of the separating fragments. The present experiment furnishes additional information concerning each of the two stages of the process envisioned; in particular, the yields of the particle types newly observed, which are pertinent to the release stage, and their energy spectra, pertinent to both stages.

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[22] R. A. Nobles, Phys. Rev. 126, 1508 (1962).

TABLE I. Observed yield of light charged particles spontaneously emitted from Cf^{252}.

Particle type (x)	Energy interval observed $\Delta E(x)$, MeV	Number identified	Number per total He^4 [a]: In energy interval observed $\sum' N(x)/\sum N(He^4)$ [b]	Number per total He^4 [a]: Estimated in total energy interval $\sum N(x)/\sum N(He^4)$ [c]	Number per fission [d]
H^1	3.18–19.62	3785	$5.0 \pm 0.1 \times 10^{-2}$	$>5.0 \times 10^{-2}$	$>1.6 \times 10^{-4}$
(H^1)				$1.6 \pm 0.2 \times 10^{-2}$	$5.1 \pm 0.5 \times 10^{-5}$ [e]
H^2	3.83–18.22	462	$5.8 \pm 0.3 \times 10^{-3}$	$6.3 \pm 0.3 \times 10^{-3}$	$2.0 \pm 0.1 \times 10^{-5}$
H^3	3.89–23.13	8096	$5.5 \pm 0.2 \times 10^{-2}$	$5.9 \pm 0.2 \times 10^{-2}$	$1.90 \pm 0.06 \times 10^{-4}$
He^3	10.75–33.75	450	$\lesssim 8.2 \pm 0.4 \times 10^{-3}$	$\lesssim 9 \times 10^{-3}$	$\lesssim 2.9 \times 10^{-5}$ [f]
He^4	7.75–34.75	96 099	9.36×10^{-1}	1.00×10^{0}	$3.27 \pm 0.10 \times 10^{-3}$
He^6	10.75–33.25	1031	$1.81 \pm 0.06 \times 10^{-2}$	$2.4 \pm 0.5 \times 10^{-2}$	$7.8 \pm 1.6 \times 10^{-5}$
He^8	12.0 –36.0	31	$9 \pm 1.6 \times 10^{-4}$	$1.8 \pm 0.5 \times 10^{-3}$	$5.9 \pm 1.6 \times 10^{-6}$
He^{10}	13.0 –36.0	2	$5 \pm 4 \times 10^{-5}$	$1 \pm 1 \times 10^{-4}$	$3 \pm 3 \times 10^{-7}$
Li	19.0 –39.0	23	$6 \pm 1.3 \times 10^{-4}$	$1.2 \pm 0.6 \times 10^{-3}$	$3.9 \pm 2.0 \times 10^{-6}$
Be	33.0 –41.0	3	$1 \pm 0.6 \times 10^{-4}$	$>1 \times 10^{-4}$	$>3 \times 10^{-7}$

[a] The total number of He^4, $\Sigma N(He^4)$, produced in a run was obtained by extrapolation of the observed spectrum to zero energy using the spectral shape measured by Nobles (Ref. 22).

[b] $\Sigma' N(x)$ is the sum over the energy interval observed $\Delta E(x)$.

[c] The $\Sigma N(x)$ were estimated, where feasible, by assuming spectral shapes similar to the He^4.

[d] Based on the He^4 yield reviewed by T. D. Thomas and S. L. Whetstone, Jr., Phys. Rev. **144**, 1060 (1966).

[e] Yield of the component of the spectrum that peaks at about 9.5 MeV, and is most likely produced in fission.

[f] The yield of He^3 is considered to be more of an upper limit due to the appreciable number of He^4 events undoubtedly dispersed into the neighborhood of the He^3 locus.

A. Yields of the Light Charged Particles

Halpern[2,3,23] has estimated the amount of energy $E_R(x)$ required for the release of a particle of type x into the region between the two fragments at the time of scission, taking into account the separation energy of the particle x from one of the fragments, the change in Coulomb potential energy of the particle when removed from one of the fragments and placed midway between the fragments, and the average kinetic energy of the particle at the time of its release.

We have calculated $E_R(x)$ for all particles of charge number $Z \leq 4$ that are included in the current Chart of the Nuclides[24]; the results are summarized in Table II. The release energies are found to be somewhat smaller, with one marginal exception, for separation from the heavy member of the fragment pair, this trend being more pronounced for the more massive emitted particles. This suggests that at least the heavier emitted particles may actually be emitted predominantly at the expense of the heavy fragments, as was noted previously by Feather[25] for alpha-particle emission. This would be

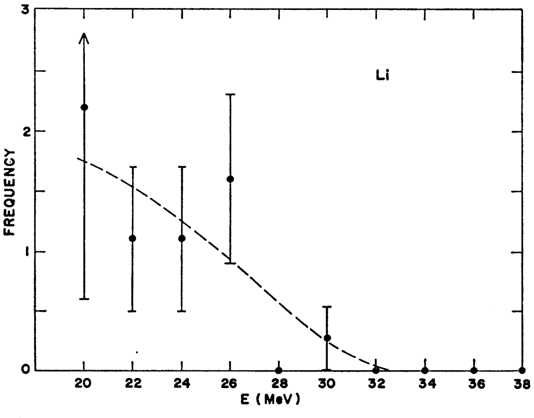

FIG. 7. Energy spectra for the particles with $Z=3$ corrected for loss in the aluminum absorber foil.

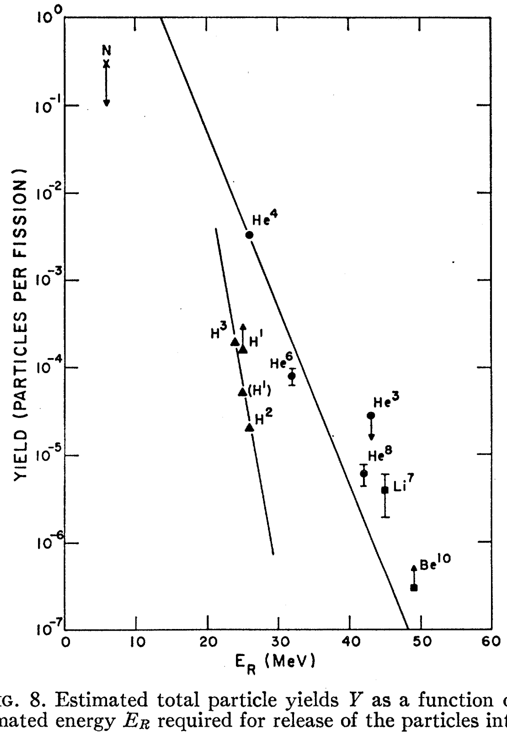

FIG. 8. Estimated total particle yields Y as a function of the estimated energy E_R required for release of the particles into the space between the fission fragments.

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[23] I. Halpern (private communication) suggests now that K is smaller than previously indicated for He^4 emission, and, moreover, nearly the same for particles of all masses.

[24] *Chart of the Nuclides* (Knolls Atomic Power Laboratory, General Electric Company, Schenectady, New York, 1964), 7th ed.

[25] N. Feather, Nature **159**, 607 (1947); Proc. Roy. Soc. (Edinburgh) **A46**, 192 (1964).

TABLE II. Light particles emitted in spontaneous fission of Cf^{252}.

	$x^{\rm a}$	$T_{1/2}{}^{\rm b}$	$E_R(L)^{\rm c}$ (MeV)	$E_R(H)$ (MeV)	$Y^{\rm d}$	Reference
1	n	12 min	7	6	$\lesssim 3\times10^{-1}$	e
2	H^3	12.26 yr	27	24	$(1.90\pm0.06)\times10^{-4}$	f
3	H^1	∞	24	25	$>1.6\times10^{-4}$	f
					$(5.1\ \pm0.5)\ \times10^{-5}$	f, g
4	H^2	∞	27	26	$(2.0\ \pm0.1)\ \times10^{-5}$	f
5	He^4	∞	29	26	$(3.27\pm0.01)\times10^{-3}$	f
6	He^5	2×10^{-21} sec	34	30	*	h
7	He^6	0.81 sec	39	32	$(7.8\ \pm1.6)\ \times10^{-5}$	f
8	He^7	10^{-21} sec	50	41	*	h
9	He^8	1.22×10^{-1} sec	52	42	$(5.9\ \pm1.6)\ \times10^{-6}$	f
10	He^3	∞	43	43	$\lesssim 2.9\times10^{-5}$	f
11	Li^7	∞	51	45	$(3.9\ \pm2.0)\ \times10^{-6}$	f, i
12	Li^8	0.85 sec	53	46		i, j
13	Li^9	0.17 sec	56	47		i
14	Li^6	∞	52	48		i
15	Be^{10}	2.7×10^6 yr	57	49	$>3\times10^{-7}$	i, k
16	Be^8	$\sim3\times10^{-16}$ sec	56	50	*	h, i, j
17	Li^5	$\sim10^{-21}$ sec	53	51	*	h, i
18	Be^9	∞	58	52		i
19	Be^{11}	13.6 sec	61	53		i
20	Be^7	53 day	69	66		i, l
21	Be^6	$\gtrsim4\times10^{-21}$ sec	76	74	*	h, i
...	B^{11}	∞	68	63		m

[a] x: Particle type; includes all nuclides with $Z\leq 4$ as given in the *Chart of the Nuclides* (Ref. 24) plus He^8 and B^{11}.

[b] $T_{1/2}$: Half-life from *Chart of the Nuclides* (Ref. 24) except for He^7 from C. Détraz *et al.*, Phys. Rev. Letters **14**, 708 (1965), and He^8 (Ref. 16).

[c] $E_R(L)$, $E_R(H)$: The energy required for release of particle "x" removed from the light (L), or heavy (H), fission fragment into the region midway between the fragments according to the Halpern model (Refs. 2, 3, and 23). $E_R=S+V+K$. S is the separation energy of the particle from one of the fragments. Mass values of the heavy nuclides from A. G. W. Cameron, U. S. Atomic Energy Commission Laboratory Report No. 433 (unpublished) and of the particles x from J. H. Mattauch, W. Thiele, and A. H. Wapstra, Nucl. Phys. **67**, 1 (1965). The tabulated release energies for separation from the light and heavy fragments are simple averages of the values calculated for the mass pairs Xe^{141}-Ru^{111}, Xe^{142}-Ru^{110}, Xe^{143}-Ru^{109}, I^{142}-Rh^{110}, and Cs^{142}-Tc^{110} which are believed to be representative of the primary fission fragments that ultimately produce the maximum observed radiochemical yield (Ba^{140},6.3%) reported by W. E. Nervik, Phys. Rev. **119**, 1685 (1960). V is the Coulomb potential energy change to attain the assumed release configuration. $V(L)=zV_0(1+Z/Z_L)/(Z-Z_L)$. $V(H)=zV_0(1+Z/Z_H)/(Z-Z_H)$. z is the charge number of particle. Z, Z_L, and Z_H are the charge numbers of the fissioning nucleus, the light, and the heavy fragments (98, $\approx$44, $\approx$54). V_0 is the Coulomb potential energy of fragments at scission time, assumed also to be time of particle release ($\approx$165 MeV). K is the average kinetic energy of the particle at the time of release ($\approx$2 MeV).

[d] Y: The observed total number of particles per fission.

[e] Estimated yield of "scission" neutrons from H. R. Bowman, S. G. Thompson, J. C. D. Milton, and W. J. Swiatecki, Phys. Rev. **126**, 2120 (1962).

[f] From Table I, this paper.

[g] Yield given is for partial yield (H^1). See footnote (d), Table I.

[h] Asterisks denote nuclides with half-lives too short to permit identification in this experiment.

[i] Yields given after Li^7 and Be^{10} are for all Li or all Be, respectively.

[j] Yield of Li^8-Be^8: $<1\times10^{-5}(Cf^{252})$, M. L. Muga, H. R. Bowman, and S. G. Thompson, Phys. Rev. **121**, 270 (1961).

[k] Yield of Be^{10}: $<4\times10^{-6}(U^{235}+n_{\rm th})$, K. F. Flynn, L. E. Glendenin, and E. P. Steinberg, Phys. Rev. **101**, 1492 (1956).

[l] Yield of Be^7: $<3\times10^{-9}(U^{235}+n_{\rm th})$, J. C. Roy, Can. J. Phys. **39**, 315 (1961).

[m] B^{11} has the lowest calculated E_R of the $Z=5$ nuclides.

in agreement with a simple interpretation of the fragment mass-yield results of Schmitt, Neiler, Walter, and Chetham-Strode.[26] The observed yields of the various particles are included in Table II; the general anticorrelation of these yields and the calculated release energies confirms the conclusions reached earlier by Halpern.[2,3]

Although, as Halpern[2,3] has emphasized, it does not seem possible for most of these particles to be released in a conventional "evaporation" process, it might still be expected on general grounds that the yields of the various particles should depend on the energy required for their release according to $\exp[-E_R/T]$, where the parameter T would correspond crudely to a nuclear temperature. A plot of the observed particle yields on a logarithmic scale versus the release energy E_R for separation from the heavy fission fragment is shown in Fig. 8. The straight lines that can be drawn through the yields of the $Z=1$ and $Z=2$ particles support to some extent the validity of the exponential dependence and the estimates of E_R. Values of 0.9 and 2.2 MeV are found for the parameter T, for the $Z=1$ and $Z=2$ particles, respectively, not too different from the 1-MeV value commonly found for the nuclear temperature. In either case, however, a yield of scission neutrons much larger than believed possible would be predicted by extrapolation of these lines.

Our calculation of the Coulomb potential energies has assumed a fragment mutual Coulomb potential energy at the time of particle release of 165 MeV (see Table II). This value[27] is obtained from the measured average value of the total fragment kinetic energy ($\sim$185 MeV) in the normal spontaneous fission of Cf^{252}, reduced by $\sim$20 MeV to allow for the energetic alpha-particle emission. If a smaller value of V_0 is used, corresponding to a greater average separation distance of the fragments at scission in the case of light charged particle emission, the contribution of the Coulomb energy to the energy required for release would be reduced in proportion to the charge number of the emitted particle. The $Z=1$ and $Z=2$ data would then tend to group more closely together on the plot of Fig. 8.

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

[27] H. W. Schmitt, W. E. Kiker, and C. W. Williams, Phys. Rev. **137**, B837 (1965); S. L. Whetstone, Jr., *ibid.* **131**, 1232 (1963).

B. Energy Spectra of the Light Charged Particles

In the second stage, which succeeds the release of the particles, the Coulomb repulsions between the particles and the fragments largely determine the distributions in final energy and angle, although the time and place of release and the initial kinetic energy also have important effects.[2]

The energy spectra observed for the charge $Z=2$ particles show quite clearly for He^3, He^4, and He^6, and plausibly for He^8 the same general shape, a broad peak (8–12 MeV wide) which reaches its maximum at about 15 MeV. On closer examination it is found that in order of increasing mass the spectra peak at 17 ± 1, 16 ± 0.5, 13 ± 1, and $\lesssim13$ MeV, respectively. This inverse dependence of particle energy on particle mass can be the result of a number of factors.

First, on the simplest assumption that the average energy of emission is the same for all particles of $Z=2$, conservation of momentum will, in going from He^3 to He^8 emission, increase the amount of energy given to the recoiling fragment system, and decrease the energy of the light particles, but only by about 2%, or some 0.3 MeV, a change an order of magnitude too small to account for our observations.

On the basis of Halpern's model[2] we can make a qualitative estimate of the effect of changing the mass of the emitted particle. The particle is being accelerated by a potential decreasing with time. A very light particle will be quickly accelerated and will acquire a kinetic energy more or less equal to the potential at time $t=0$; a heavier particle, accelerated more slowly, will acquire less kinetic energy. Similarly, if two particles have different initial velocities, the one with the higher velocity will receive more energy from the potential than will that with lower velocity. On both of these counts we may expect the He^6 particle to have a lower final energy than the He^4.

We have made a quantitative estimate of this effect by applying some simplifying approximations to Halpern's model.[2] The two fission fragments are taken to be both of $Z=49$ and $A=126$ and separate from one another as if they alone provided the entire Coulomb potential. The initial separation of charges is taken to be 20.5×10^{-13} cm (after Halpern).[2] The light charged particle is released at some time t after scission and the beginning of the fragment motion at a point midway between the two fragments, with some initial kinetic energy K, and a direction of motion perpendicular to the line between the two fragments. The time-dependent potential felt by the third particle can be easily calculated, and its subsequent motion determined by numerical integration. Because of the uncertainty concerning the starting conditions, we have done these calculations on the basis of three sets of assumptions: (1) The initial particle energy is $17.6/m$ MeV, where m is the particle mass in amu, and the release time is 1.1×10^{-21} sec. These are the conditions originally suggested by Halpern.[2,3] (2) The initial energy is $6.4/m$ MeV and the release time is 0.6×10^{-21} sec. This energy would seem to be reasonable for the lower mass particles, and the release time is chosen to make the final alpha-particle energy close to 16 MeV. (3) The initial energy is 1.6 MeV and the release time is 0.6×10^{-21} sec. This assumption is to investigate the consequences of Halpern's recent suggestion[23] that the initial energy is more or less independent of mass. The results of these calculations are summarized in Table III. We note that regardless of assumptions about initial conditions, the calculations produce a shift between the kinetic energies of the various helium ions that is in accord with the experimental data. Considering the crudeness of the model used, however, one cannot really decide between the three sets of assumptions. The calculated energies for the charge-one particles are all higher than the most probable observed energies. In order to obtain agreement between calculation and experiment, we will need to use either a lower initial kinetic energy or a later release time. The assumption of an inverse mass dependence of the initial energy will further increase ths differences between the calculated numbers, whereas experimentally the most probable values of the energy are more nearly the same for the charge-one particles.

TABLE III. Results (in MeV) of calculation of final kinetic energies of various particles.

Initial energy Release time	$(17.6/m)$ MeV 1.1×10^{-21} sec	$(6.4/m)$ MeV 0.6×10^{-21} sec	1.6 MeV 0.6×10^{-21} sec	Experimentally observed most probable energies (MeV)
H^1			11.4	9 ± 2
H^2			10.0	7 ± 2
H^3			9.0	8 ± 1
He^3	20.3	19.1	17.8	17 ± 1
He^4	17.1	16.2	16.2	16 ± 0.5
He^6	12.6	11.8	13.8	13 ± 1
He^8	9.5	8.5	11.7	$\lesssim13$

Other effects may also contribute to the shift in most probable energy that is seen for the helium isotopes. One might expect those particles that require larger amounts of release energy E_R to be produced only in the fissions of larger distortion—in the so-called "late" fissions.[2] In this case, for example, the He^6 particles would be expected to begin their acceleration in a lower Coulomb potential than do the more easily produced He^4 particles.

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