

Total Energy Associated with Prompt γ -Ray Emission in the Spontaneous Fission of ^{252}Cf

E. Nardi

*Soreq Nuclear Research Center, Yavne, Israel,
and The Weizmann Institute of Science, Rehovot, Israel*

and

A. Gavron and Z. Fraenkel

*The Weizmann Institute of Science, Rehovot, Israel
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The total energy associated with the emission of prompt γ rays in spontaneous fission of ^{252}Cf was measured as a function of the fission-fragment mass and kinetic energy. The γ -ray energy shows a saw-tooth curve behavior as a function of mass, and a small decrease with increasing kinetic energy. The measurements were carried out using plastic scintillators.

I. INTRODUCTION

The study of total γ -ray energy in fission is necessary in order to obtain information regarding the final steps of the deexcitation process of fission fragments. Particularly, one may be able to ascertain whether angular momentum effects play a role in fission-fragment deexcitation. High values of angular momentum could cause γ -ray emission to compete favorably with neutron emission thereby causing an increase in the total γ -ray energy release. In addition to this, the knowledge of the total γ -ray energy is necessary when considering total energy release in fission.

Pleasanton, Ferguson, and Schmitt¹ recently measured the total γ -ray energy release as a function of fragment mass and kinetic energy in the thermal neutron-induced fission of ^{235}U . The γ rays were measured using a NaI detector and the total γ -ray energy was obtained by unfolding the experimental data. As will be seen in the following, our results obtained for ^{252}Cf are similar to the results of Pleasanton, Ferguson, and Schmitt for ^{235}U .

Recent work on the total γ -ray energy in ^{252}Cf was reported by Nifenecker *et al.*² and Verbinsky, Weber, and Sund.³ The former authors measured the total γ -ray energy as a function of mass ratio and fragment kinetic energy, but not as a function of the individual fragment masses. The latter authors obtained the average total γ -ray energy release in ^{252}Cf . We also mention the spectroscopic studies of prompt γ -ray emission in fission using high-resolution GeLi detectors in which much spectroscopic information (especially in even-even fission fragments) was obtained.⁴

We have measured the total energy emitted as γ rays in spontaneous fission of ^{252}Cf , as a function of the fission-fragment mass and total kinetic

energy. The measurements were performed using plastic scintillators, which enabled us to determine the total γ -ray energy release without invoking unfolding procedures. In the following we present a summary of the experimental method and present our results and conclude with a discussion of their significance.

II. EXPERIMENTAL METHOD

In a previous publication⁵ it was shown that the total γ -ray energy absorbed by a plastic scintillator is to a good approximation proportional to the total γ -ray energy incident on it. We have used this property to measure the total γ -ray energy released by fission fragments in the fission of ^{252}Cf .

Assume a γ -ray source emits N_0 photons of energy E_0 . We denote $N(E)$ as the number of counts of energy E detected due to this source in a plastic scintillator. We define $\eta(E_0)$, the ratio between the total absorbed and emitted energies, as follows:

$$\eta(E_0) = (N_0 E_0)^{-1} \int_{E_m}^{E_0} N(E) E dE, \quad (1)$$

where E_m is the minimum detectable γ -ray energy in the spectrometer system. Our measurement is based on the assumption that $\eta(E_0)$ is almost independent of E_0 . The quantity $\eta(E_0)$ was calculated by the method described in Ref. (5) and was obtained experimentally using a set of calibrated monoenergetic γ -ray sources (see below).

The γ -ray energy emitted by each of the fission fragments was determined using the method of Maier-Leibnitz, Schmitt, and Armbruster.⁶ The energy of photons emitted by fragments moving towards a scintillator increases due to the Doppler effect and their detection probability increases due to the increase in the effective solid angle of the

detector. This gives rise to an anisotropy of the measured γ -ray energy, depending on the fragment velocity and direction. We denote $E_{\gamma i}$ as the total γ -ray energy emitted by a fragment mass A_i ($i=1,2$; $A_1=252-A_2$), E_γ as the total energy emitted by the two fragments, and e_i as the measured γ -ray energy for a fragment with mass A_i moving in the direction of the detector. The asymmetry parameter Δ is defined by $\Delta=(e_1-e_2)/(e_1+e_2)$. Denoting β_i as the velocity of the fragment i (in units of c) and θ as the angle between the direction of the fission fragments and the γ rays, we have to first order in β^6 :

$$\frac{E_{\gamma 1}}{E_\gamma} = \frac{A_1}{252} \left(1 + \frac{\Delta}{3\beta_2 \cos \theta} \right). \quad (2)$$

Our experimental arrangement is shown in Fig. 1. A thin ^{252}Cf source of approximately 10^5 fissions/min was placed in an aluminum vacuum chamber between two surface-barrier fission detectors, each subtending an angle of 24° with respect to the source. The vacuum chamber was 30 cm in diameter and had a wall thickness of 0.5 cm. Two identical NE102A plastic scintillators denoted PM1 and PM2 of 12.5-cm diameter and 5.0-cm length, mounted on 58 AVP photomultipliers, were placed outside the chamber on both sides of the source at a distance of 60 cm from the source. The electronic system associated with PM1 was calibrated to measure the energy spectrum in the 25–210 keV range, whereas the system associated with PM2 was calibrated to measure energies between 210 keV and 7 MeV. γ rays were separated from neutrons by means of the time-of-flight method. The width (full width at half maximum) of the time-of-flight γ -ray peak was 1.5 nsec; the prompt γ peak was completely separated from the neutron time-of-flight spectrum.

The γ -ray energy vs pulse-height calibration curves were determined separately for PM1 and

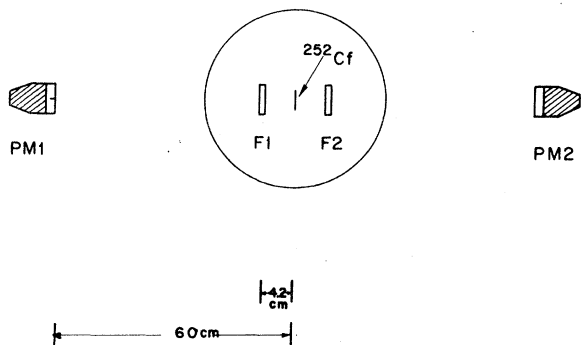


FIG. 1. Schematic description of experiment. F1 and F2 denote the solid-state detectors while PM1 and PM2 are the plastic scintillators.

PM2, using the method of Ref. 5. This method involves a comparison of the ^{203}Hg , ^{113}Sn , ^{103}Ru , ^{137}Cs , ^{54}Mn , and ^{60}Co experimental spectra with results of Monte Carlo calculations.⁵ As defined in Eq. (1), $\eta(E_0)$ was derived by taking in the numerator the sum of $\int_{0.025\text{MeV}}^{0.211\text{MeV}} N(E)E dE$ as recorded by PM1 and of $\int_{0.211\text{MeV}}^{E_0} N(E)E dE$ as measured by PM2. The result is shown in Fig. 2. The $\eta(E_0)$ vs E_0 curve is seen to be constant above 0.7 MeV. At 0.5 MeV however, η is observed to increase by about 10% while at 0.3 MeV the value of η is 5% lower than η above 0.7 MeV. Monte Carlo calculations show that the $\eta(E_0)$ curve is flat above 1 MeV.⁵ Therefore these results can be extrapolated to much higher energies. The masses and kinetic energies of the fission fragments were determined using the calibration method of Schmitt, Kiker, and Williams⁷ and an iteration procedure similar to that of Watson *et al.*⁸ The mass resolution was of the order of 5 amu and the total kinetic-energy resolution was roughly 7 MeV.⁹

Three-dimensional events consisting of the two fission-fragment energies and the γ -ray pulse height in either PM 1 and PM2 were stored on magnetic tape. Data were recorded at the angles $\theta=0$ and 60° . This was done in order to check the consistency of our results at two angles. A total of about 750 000 three-dimensional events were stored and processed.

III. RESULTS

The average total γ -ray energy release, averaged over all the fission-fragment mass ratios, was found [using Eq. (1)] to be equal to

$$\bar{E}_{\gamma T} = 6.7 \pm 0.4 \text{ MeV}.$$

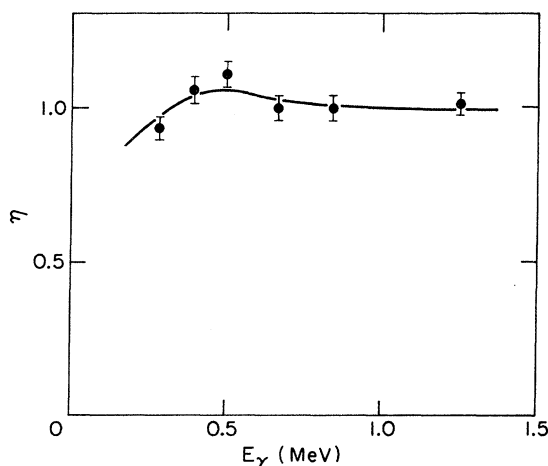


FIG. 2. η as a function of the γ -ray energy. The points are results obtained using the calibrated sources of ^{203}Hg , ^{113}Sn , ^{103}Ru , ^{137}Cs , ^{54}Mn , and ^{60}Co .

The error quoted includes the error due to the variation in $\eta(E_0)$ over the entire energy range. This result is in good agreement with the value reported by Verbinski, Weber, and Sund³ who obtained 6.84 MeV for ²⁵²Cf. Our result is substantially lower than the values of 8.2 and 9.0 MeV quoted by Maier-Leibnitz, Armbruster, and Specht¹⁰ for ²⁵²Cf.

The average γ -ray energy emitted by a fragment $\bar{E}_\gamma(A, E_k)$, as a function of the mass A and total kinetic energy E_k , was obtained by using Eq. (2). No systematic difference was observed between results of \bar{E}_γ obtained at $\theta=0$ and $\theta=60^\circ$ beyond what could be attributed to the various systematic errors in our method of analysis.

In Fig. 3 are plotted the values of $\bar{E}_\gamma(A)$ vs A at 0° . The error bars in Fig. 3 are due to statistical errors only and do not include the systematic error due to the relative deviations of $\eta(E_0)$ from a constant value. This graph shows a "saw-tooth" structure similar in shape but less pronounced than the well known neutron "saw-tooth" curve. A similar result was obtained by Pleasonton, Ferguson, and Schmitt¹ in the thermal neutron fission of ²³⁵U. These authors also used the method proposed by Maier-Leibnitz, Schmitt, and Armbruster⁶ [see Eq. (3)] but measured the γ rays with

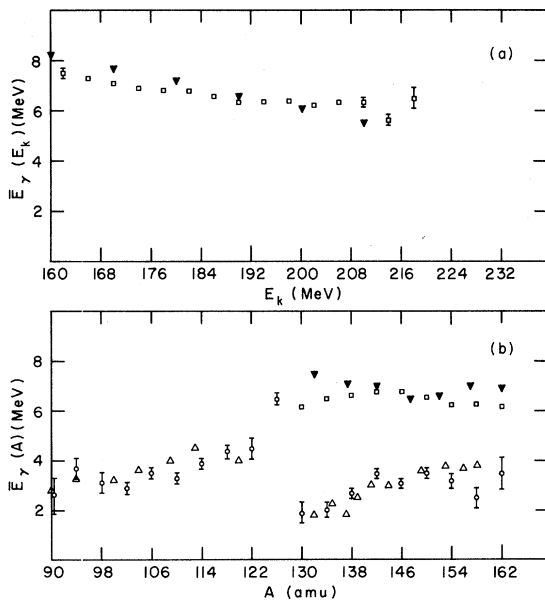


FIG. 3. (a) $\bar{E}_{\gamma T}$ as a function of E_k . The squares are from the present experiment and the full triangles are data obtained by Nifenecker *et al.* (Ref. 2). (b) \bar{E}_γ as a function of A (the fission-fragment mass), given by the circles. The squares are $\bar{E}_{\gamma T}$ as a function of the mass ratio and the full triangles are the corresponding data obtained by Nifenecker *et al.* (Ref. 2). The open triangles are the calculated values of $\bar{E}_\gamma(A)$ taken from Ref. 12.

a NaI crystal. They obtained the initial γ -ray spectrum from the experimental spectrum using a weighting method based on the response of a NaI scintillator to individual γ rays.

We also show in Fig. 3 the total γ -ray energy (emitted by the two complementary fragments) $\bar{E}_{\gamma T}$ as a function of the mass of the heavy fragment. Similar results obtained recently by Nifenecker *et al.*² are also shown for comparison. The differences are probably due to the systematic errors in the different experimental method used. The measurements of Nifenecker were performed with a large liquid scintillator tank. Background interference due to proton recoils by neutrons was subtracted event by event by counting the number of neutrons after thermalization, whereas our method separates prompt γ rays from neutrons by the time-of-flight method. The measurement of Nifenecker *et al.* was over a 4π solid angle, whereas our measurements were performed with low geometric efficiency at only two angles of 0 and 60° . Hence, Nifenecker *et al.* could not measure the total γ -ray energy release from individual fission fragments.

In Fig. 3 are plotted $\bar{E}_{\gamma T}$ as a function of E_k , together with the values obtained by Nifenecker *et al.* Our values of $\bar{E}_{\gamma T}$ change less with E_k than those of Nifenecker and resemble those of Pleasonton, Ferguson, and Schmitt¹ for ²³⁵U.

In Fig. 4 we have plotted the average energy recorded by the plastic scintillator per detected γ ray, $\bar{\epsilon}_\gamma$, as a function of the mass ratio of the fission fragments. Although $\bar{\epsilon}_\gamma$ is not equal to the average energy of the primary γ rays, it is reasonable to assume that $\bar{\epsilon}_\gamma$ vs (A_L, A_H) reproduces the trend of the average primary photon energy as

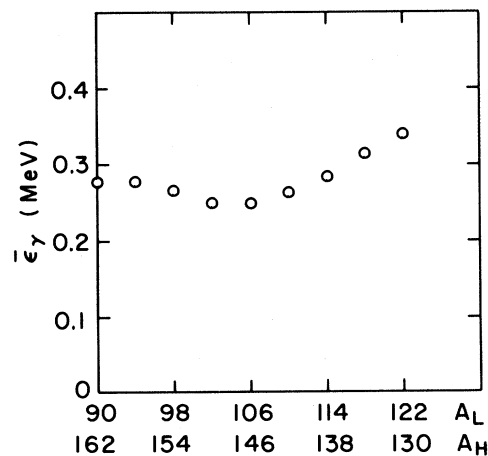


FIG. 4. Average energy, $\bar{\epsilon}_\gamma$, deposited by γ rays in the plastic scintillator as a function of the mass ratio. The errors are too small to be shown in the graph.

a function of mass ratio. The highest values of $\bar{\epsilon}_\gamma$ were obtained for the mass division 118, 134 and 122, 130 (i.e., in the region of the closed shells of $Z=50$ and $N=82$ for the heavy fragment). The nuclear temperature is known to be relatively high in the region of closed shells, hence the average photon energy in the above mentioned mass intervals is higher than for the other mass regions.¹¹

IV. DISCUSSION

We have seen that the variation of \bar{E}_γ as a function of A and E_k resembles the behavior of the average neutron number $\bar{\nu}$ as a function of these variables. Two effects can explain this behavior: Variation of the neutron binding energy of the residual fragments as a function of A and E_k and variation of the angular momentum of the primary fission fragments as a function of A and E_k . We now proceed to show that most of the variation of \bar{E}_γ can be explained chiefly by the former effect. First, we consider $\bar{E}_\gamma(A)$ presented in Fig. 3. The open triangles are the results of an evaporation calculation.¹² In this calculation it was assumed that the angular momentum does not change as a result of neutron emission. Hence the effects of angular momentum in the evaporation cascade were ignored. We see that the experimental results fit the values obtained by the evaporation calculations rather well.

We now turn to interpret the dependence of \bar{E}_γ on the total kinetic energy E_k . The average experimental value of the derivative of \bar{E}_γ with respect to E_k , $\langle \partial \bar{E}_\gamma / \partial E_k \rangle$ (averaged over A and E_k), is 0.036 ± 0.03 . From the mass tables of Garvey *et al.*¹³ we find that every neutron emitted causes an average increase in the neutron binding energies of approximately 0.26 MeV. (We assume that the charge distribution at a given mass is independent of the kinetic energy and average over neighboring masses as in Ref. 12.) We have found elsewhere¹⁴ that a decrease of E_k by about 7 MeV causes an increase in $\bar{\nu}_T$ (the average total number

of neutrons emitted) by one neutron. Thus $\Delta E_k = 7$ MeV causes an additional neutron to be emitted from *one* of the fragments. The binding energy of the next neutron in this fragment will be increased by 0.26 MeV on the average. Since the γ -ray energy is approximately equal to $BE/2$, this increase in BE will result in an additional $\Delta E_\gamma = 0.13$ MeV of γ -ray energy. Therefore the calculated value of $\langle \partial \bar{E}_\gamma / \partial E_k \rangle$ is $0.13/7 \cong 0.02$. Comparing this to the experimental value of $\langle \partial \bar{E}_\gamma / \partial E_k \rangle_{\text{exp}} = 0.036$ we see that the experimental value is larger by approximately a factor of 2. The difference could be due to angular momentum effects. From calculations of Thomas and Grover¹⁵ we estimate that an increase in γ -ray energy of the order of 0.2 MeV can result from an increase of one unit of angular momentum due to the increase in the height of the Yrast level. Thus if we attribute the 0.02 difference to angular momentum effects, it implies an increase of the fragment spin by about one unit for every 10 MeV of total kinetic energy. We must however regard this estimate with caution since the theoretical value of $\langle \partial \bar{E}_\gamma / \partial E_k \rangle$ can be in error if the charge division depends significantly on E_k . In addition, we are using the mass tables for mass values extrapolated 2–3 mass units from the nearest experimental values. We thus consider the value of one unit of spin per 10 MeV kinetic energy, an order-of-magnitude estimate of the maximum possible fragment spin dependence on the total kinetic energy. Our estimate is compatible with the results of Wilhelmy *et al.*¹⁶ who found that J is independent of E_k within ± 1 units of J and with Pleasonton, Ferguson, and Schmitt¹ who also obtain a very small $\bar{E}_\gamma(E_k)$ dependence. On the other hand, Nifenecker *et al.*² find a large dependence of J on E_k . They obtain $\langle \partial E_\gamma / \partial E_k \rangle \cong 0.1$ which can be explained only if a large $J(E_k)$ dependence is assumed. It is possible that the discrepancy is due to the different experimental methods. We recall that Nifenecker *et al.*² subtracted the background due to proton recoils while in our experiment this was not necessary.

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