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Abstract

The energy spectra, multiplicities and average energies of the prompt, total and delayed γ-rays accompanying the spontaneous fission of ^{252}Cf were collected from the literature and dedicated databases. They were carefully analysed for consistency with a view to producing reference data for usage in various applications. This could be accomplished for the prompt fission gamma ray spectrum up to 20 MeV since dozens of measurements exist and reasonably agree. The prompt fission gamma ray spectrum (PFGS) was non-model evaluated by fitting the preselected experimental data with the help of the generalized least-squares (GLS) code GMA. The resulting spectrum could be considered as a reference for the γ-ray energies from 0.1 to 20 MeV with uncertainties varying between ≈ 3 and 25%. This reference gamma spectrum will be a substantial contribution to the precise and complete characterisation of the ^{252}Cf source since the prompt fission neutron spectrum (PFNS), which has been accepted as a standard for a long time, has comparable uncertainties. The average gamma multiplicity and energy were also surveyed and used to derive the recommended values. The prompt X- and γ-ray energy spectra below ≈ 100 keV and delayed photon spectra in the whole energy range, as well as their multiplicities, are still seldomly and incompletely measured, that excepts an evaluation based on experimental data. The comparison with existing theoretical prompt and delayed ^{252}Cf(s.f.) γ-spectra or with those presented in the major evaluated cross section libraries explored their incompleteness or deviations from the evaluated PFGS. The existing measurements of the pionic and muonic radioactivity of ^{252}Cf(s.f.) and ^{235}U(n_{th},f) were reviewed and the potential impact of gammas from the π^0 decay on the high energy part of the PFGS was investigated.

February 2024

Content

1. Introduction

The ^{252}Cf source is widely used in various fundamental and practical applications. The prompt fission neutron energy spectrum (PFNS) from the spontaneous fission of ^{252}Cf is one of the well-known nuclear characteristics. It was evaluated with high accuracy and was included in the neutron cross section Standards [1], [2]. Given this knowledge of the neutron emission, the standardization of the ^{252}Cf(s.f.) γ-ray energy spectrum presents a valuable task. The dominance of gamma emission over the neutrons is an additional stimulating reason: every ^{252}Cf spontaneous fission event emits around six gammas per each single neutron.

The prompt and total ^{252}Cf(s.f.) γ-ray spectra have been measured in many independent experiments since 1956. The various detectors and measuring techniques used increase the reliability of the accumulated experimental data. Practically all of them are already compiled in the EXFOR data base [3], which can thus serve as a source of numerical experimental data for the present work.

As a result of the present report, the prompt fission gamma spectrum (PFGS) from ^{252}Cf(s.f.) was proposed as a reference γ-ray spectrum in the energy range from 0.1 to 20 MeV. For this purpose, the generalized least-squares (GLS) method implemented in the Gauss-Markov-Aitken (GMA) code GMA [4], [5] was used for the non-model evaluation of the PFGS. The preliminary GMA fit to the measurements has already shown that the reference prompt ^{252}Cf(s.f.)γ-ray spectra could be produced in the energy domain 0.1 - 20 MeV with uncertainty 10 - 50% [6]. The extension of reference PFGS below 100 keV and beyond 20 MeV turns out to be impossible, since the amplitude and energy shape of the γ-ray spectrum in these ranges are not well established yet, neither experimentally nor theoretically.

The emission of delayed photons from ^{252}Cf(s.f.) has comparable probability with prompt ones. Thus, the knowledge of the delayed fission gamma spectrum (DFGS), which extends up to ≈ 1.8 MeV, and multiplicity is also important for the full characterisation. The performed review of literature resources has shown that the existing relevant measurements are scarce, controversial and often report only the yields of the most intensive radiation transitions. This made it impossible to come up with recommended values.

The analysis of the major evaluated cross section libraries has shown that the prompt and delayed ^{252}Cf(s.f.)γ-rays are not represented at all or with insufficient accuracy. Thus, ENDF/B-VIII.0 [7] has no prompt ^{252}Cf(s.f.) γ-spectrum, whereas JEFF-3.3 [8] has adopted the spectrum measured in one single experiment.

To underline the practical importance of the ^{252}Cf(s.f.) gamma and neutron emission data we refer to the γ-ray leakage benchmarks with the Fe spheres driven by ^{252}Cf source used for the validation of the iron neutron-gamma transport evaluated data [6], [9].

This report is structured as follows: Section 2 briefly overviews the physically possible disintegration modes of ^{252}Cf; Section 3 summaries the status of the prompt (PFNS) and delayed (DFNS) neutron fission spectra and multiplicities for the spontaneous fission of ^{252}Cf; Section 4 collects, estimates and selects all known publications on the measured data relevant for the prompt photon spectra and averaged multiplicities; Section 5 describes the fitting method, GMA code (with applied modifications), results of the non-model evaluation of PFGS in the energy range 0.1 to 20 MeV and gamma multiplicity; Section 6 intercompares the obtained reference prompt γ ray spectrum with the corresponding information from theoretical modelling and major evaluated libraries; Section 7 deals with the status of measured data for delayed photon spectra and multiplicities; Section 8 reviews the known measurements of the pionic and muonic radioactivity of ^{252}Cf(s.f.) in comparison with $^{235}U(n_{th},f)$ and the impact of the π^0 decay gammas on PFGS at energies above 50 MeV. The report ends with a Summary, Acknowledgement and References.

2. Disintegration modes of ^{252}Cf

The disintegration of ^{252}Cf, which is accompanied by the emission of neutrons and photons (γ- and X-rays), includes several processes: prompt and delayed emission during or after spontaneous fission and after α-decay. The basic characteristics and constants of the disintegration modes of isotope ^{252}Cf and the resultant radiations are summarised in Table 2.1. The isotope ^{252}Cf decays with probability (96.914 ± 0.008)% emitting α-particles and also undergoes spontaneous fission with probability (3.086 ± 0.008)%. The corresponding half-lives are (2.6470 ± 0.0026) and (85.76 ± 0.23) years. These data are taken from the Decay Data Evaluation Project (DDEP) [10].

TABLE 2.1 Main parameters of the neutron and γ-ray radiations from disintegration of ^{252}Cf. The average multiplicities are normalized per spontaneous fission event (f), the spectrum average energies per spectrum area. References for the origin of Cf data are given.

	Neutrons			Gammas		
	Multiplicity ν, n/f	$\langle E_n \rangle$, MeV	Origin of Spectrum	Multiplicity M_γ, γ/f	$\langle \varepsilon_\gamma \rangle$, MeV	Origin of Spectrum
	Spontaneous Fission: P_{sf} = (3.086 ± 0.008)%, $T_{1/2}$ = (85.76 ± 0.23) y					
Prompt	3.7590 [7]	2.122	Standards [1]	9.194 [a)]	0.813	present work [a)]
Delayed	0.0086 [12],[7]	0.464	ENDF/B-VIII.0 [7]	10.3 [13]	0.774	Stoddard [13],[14]
Total	**3.7676** [11] [b)]			**19.49** [c)]		
	Alpha Decay: P_α = (96.914 ± 0.008)%, $T_{1/2}$ = (2.6470 ± 0.0026) y					
Total				**0.0089**	**0.070**	DDEP [10]

Note: a) see Section 5; b) see Section 3; c) arithmetic sum of prompt and delayed M_γ.

The spontaneous fission of ^{252}Cf is a source of neutrons and γ-rays. There are two emission mechanisms:

- prompt (within the first ≈ 10^{-11} - 10^{-9} s), i.e. during the fissioning of ^{252}Cf and fast de-excitation of the primary fission products;
- delayed (decay time period > 10^{-9} s), i.e. de-excitation of the isomer states in the primary fission fragments (FF) and de-excitation of residuals after β^- decay of the neutron rich FF.

Alpha decay of ^{252}Cf is an additional source of γ-rays. This process populates the ground state of ^{248}Cm with probability 81.7% and its excited levels with 15.33% [10]. The de-excitation of these levels proceeds most probably via internal electron conversion, but also with a low emission rate of the discrete γ-rays with energies (and probabilities per ^{252}Cf disintegration): 43.40 keV (0.0152%), 100.2 keV (0.0119%) and 154.5 keV (0.00051%) [10]. The γ-ray multiplicity $M_\gamma(\alpha)$ caused by alpha decay being normalized per one neutron from ^{252}Cf(s,f.) is equal to 0.0089 γ/f or only 0.043% of the total spontaneous fission γ-multiplicity M_γ. Due to this, the gammas from $^{252}Cf(\alpha)^{248}Cm$ could be excluded from consideration.

It is worth mentioning that the maximum energy of γ-rays emitted from Cf(s.f.) in the prompt mode cannot exceed the total fragment kinetic energies before neutron emission, which was experimentally found to be 186.5 ± 1.2 MeV [15].

The sources of delayed γ-rays are de-excitation of isomeric states in the fission products and gammas which follow betta-decay. The maximum energy of delayed gamma rays, as experimentally observed, is around 1.8 MeV (see Section 7). This means that prompt and total (i.e. the sum of prompt and delayed) γ-ray emission spectra should be identical above this energy.

^{252}Cf emits neutral and charged pions during fissioning. The experimentally established upper limits for this probability relative to the s.f. process amounts to ≈ 10^{-12} - 10^{-11} in case of the π^0 emission and ≈10^{-8} for $\pi^\pm$. In Section 8 more information is given and the impact of γ-rays from decay of neutral pions on the ^{252}Cf(s.f.)PFGS is discussed.

3. ^{252}Cf(s.f.) as a source of neutrons

The compact information collected in this Section about the prompt and delayed neutron energy spectra for ^{252}Cf(s.f.), their uncertainties and energy correlations, serves as a “etalon or guide”, which we would like to follow in evaluating the prompt and delayed γ-rays in the present work.

The prompt fission neutron spectrum (PFNS) from spontaneous fission of ^{252}Cf was measured in dozens of experiments. The neutron spectrum was then evaluated by W. Mannhart with help of the generalized least-squares methods in the energy range 0.1 – 15 MeV with uncertainties 1.3 – 12% [16] and was adopted as a standard physical quantity [1]. The PFNS spectrum is plotted in Fig. 3.1 in the 725 groups presentation (data are taken from the IRDFF-II database [17]). The prompt neutron multiplicity, i.e., the number of prompt neutrons per spontaneous ^{252}Cf fission is also well established in independent experiments, $\nu_p = 3.7590 \pm 0.0048$ [7].

The delayed neutron spectrum (DFNS) was taken from the ENDF/B-VIII.0 evaluation [7]. The number of delayed neutrons per spontaneous ^{252}Cf fission ν_d is equal to 0.0086 ± 0.0010 [12], [7]. These data are mainly based on the measurements.

The total fission neutron spectrum (TFNS) is obviously the sum of PFNS and DNFS. The total neutron multiplicity $\nu_{tot} = 3.7676 \pm 0.0047$ was evaluated by E. Axton in 1986 [11]. In 2018, the Standards defined from the GMA analysis the new recommended value $\nu_{tot} = 3.7637 \pm 0.0016$ (0.42%) [1]. These two values agree each other within uncertainties, the difference between them amounts only 0.10%. Because of such negligible difference for the gamma spectra evaluation and since Standards did not recommended ν_p and ν_{del} we used through the present work $\nu_{tot} = 3.7676 \pm 0.0047$.

Both spectra normalized per total neutron multiplicity $\nu_{tot} = 3.7676$ are displayed in Fig. 3.1 The fraction of delayed neutrons is relatively small $\nu_d / \nu_{tot} = 0.23\%$. However, the average delayed neutron energy $\langle E_d \rangle = 0.464$ MeV is several times less than the average energy of prompt neutrons $\langle E_p \rangle = 2.122$ MeV. Consequently, at neutron energies below 0.1 MeV the fraction of the delayed neutrons amounts already to more than 20% of the total neutron yield, Fig. 3.1

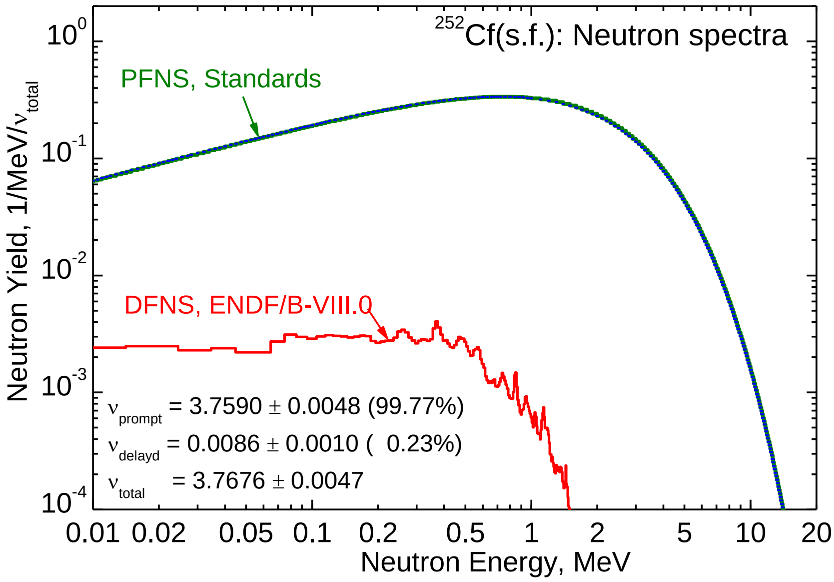

FIG. 3.1. The absolute prompt (PFNS) and delayed (DFNS) fission neutron spectra normalized per total number of neutrons for the spontaneous fission of ^{252}Cf (data are taken from IRDFF-II [17] and ENDF/B-VIII.0 [7], respectively).

The relative uncertainties for the standard PFNS of ^{252}Cf(s.f.) are displayed in Fig. 3.2. It can be seen that the minimum uncertainty 1.3 – 2.0 % is reached in the energy domain 1 to 5 MeV, then it rapidly increases up to 30% at the low energy (≤ 10 keV) and up to 80% at high energy (20 - 30 MeV) [1].

The energy-energy correlation matrix for PFNS of ^{252}Cf(s.f.) provided in Standards [1] is displayed in Fig. 3.3. It can be seen that correlations are rather small, except the neutron energy domain below 10 - 100 keV where they reach the value ≈ 0.8.

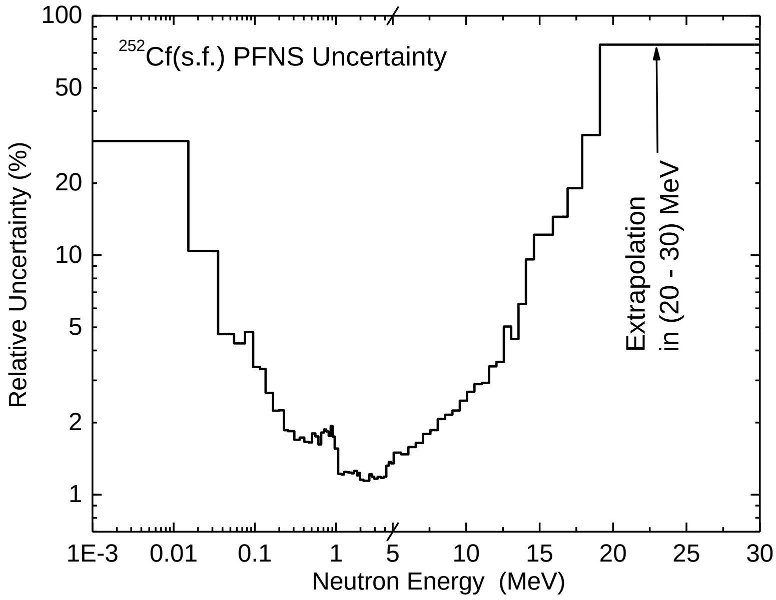

FIG. 3.2. The relative uncertainty of the prompt fission neutron spectrum (PFNS) from spontaneous fission of ^{252}Cf(s.f.) adopted in the neutron Standards [1] (data are taken from IRDFF-II [17]). Note a change of neutron energy scale at 5 MeV

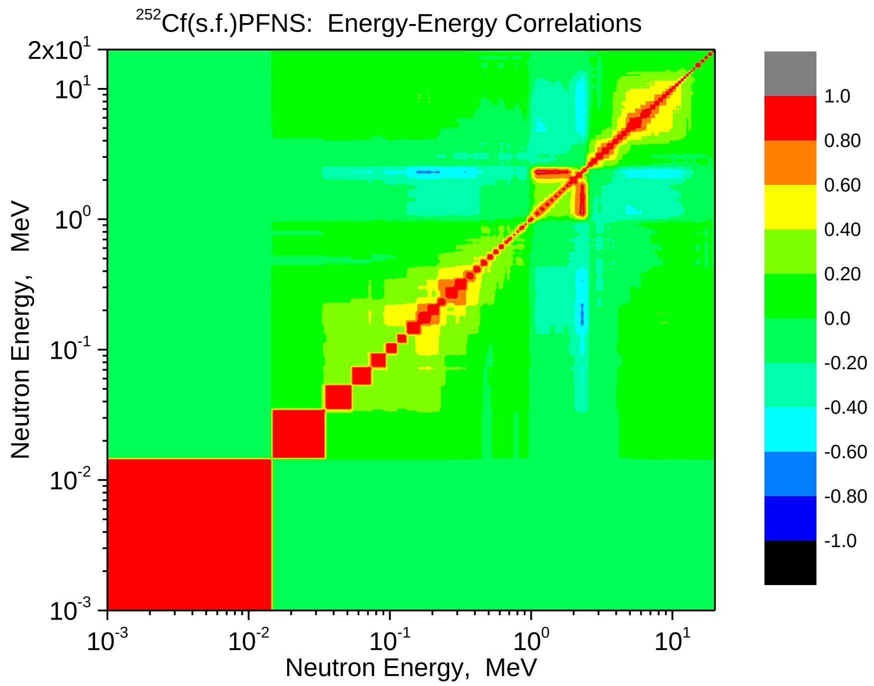

FIG. 3.3. The energy-energy correlation matrix for prompt fission neutron spectrum (PFNS) from spontaneous fission of ^{252}Cf adopted in the neutron Standards [1].

4. ^{252}Cf(s.f.) as a source of gammas

The literature and the EXFOR data base [3] contain many independent experiments carried out in different labs since 1956. Measurements were carried out with different experimental techniques and methods with the aim to observe the various characteristics of the γ- and X-ray emission from spontaneous fission of ^{252}Cf.

In this Section an overview is given of the used techniques, the measured prompt and total (i.e., prompt plus delayed) energy spectra, the average gamma multiplicities and energy. The measurements and data for gamma energies below 100 keV are analysed separately since they use different technique and are scarce. The overview includes information important for understanding the arrangement of experiments, measuring methods, applied corrections and the quality of results and their uncertainties. *Text in italics summarizes whether the reported data sets were used in the present GMA evaluation. It also contains specific details such as energy range, absolute or relative normalization, the values of uncertainties which were reported in the original publications or we have to guess them ourselves.*

4.1. Techniques used to measure the γ-ray energy spectra

The technique used to measure the Prompt Fission Gamma-ray Spectra (PFGS) from ^{252}Cf(s.f.). The relevant measuring technique employs the fast gamma-fission events (γ-f) coincidence. In other words, the time window from several (in the best experiments) to hundreds nanoseconds after the fission event moment was used as a gate within which the gammas were allowed to be registered and were considered to be prompt ones.

The Cf-source had usually a shape of a thin disc or small drop. The activity of the Cf-source ranged within (8 10^{+1} - 7 10^{+6}) f/s or (0.002 – 190) µCi, see Table 4.1. The ^{252}Cf(s.f.) fission events (f) were detected by registration of either (i) fission fragments (FF) with the help of the ionisation or gas scintillation chambers, silicon surface-barrier semiconductor (Si-SSD), parallel plate avalanche chamber (PPAC), … or (ii) prompt flash of gammas (γ) or neutrons (n) accompanying the fission event with the help of scintillators BaF_2, NE-213 viewed by the photomultiplier. Both methods provided a zero-time reference for fission events. The FF detectors often allowed counting the fission events with ≈ 100% efficiency and thus delivered the absolute values for the γ-ray spectra (absolute normalization per spontaneous fission of ^{252}Cf). The radiation flash fission event detectors provided the relative energy spectra (labelled in the present work also as "shape") since fast gammas or neutrons are counted with an efficiency less than 100% and within the effective solid angle which could not be well determined.

The energy spectra of γ-rays were measured by various types of single detectors or arrays with several or hundreds of crystals, which may cover the substantial fraction of the γ-ray emission angle. Various detecting crystals were used in these experiments: NaI, BiGeO, BaF_2, $LaCl_3$, $LaBr_3$ and others. The energy response of a single crystal may have a complicated response function to the monoenergetic γ-rays. Besides the full absorption peak, Compton scattering and escaping γ-rays produce lower amplitudes in the pulse height (PH) distribution. To suppress these events and make the response function quasi-monoenergetic, the anti-Compton shield was arranged around the main crystal in several experiments. It consisted of annual crystal surrounding the main one, which worked in an anti-coincidence regime. As a rule, the gamma detector response functions have to be measured or simulated at several energies. The measured PH distribution (apparatus spectrum) then has to be converted (unfolded) into γ-ray energy distribution.

For the detection of the γ-rays and X-rays with energy below 100 keV the drifted Si semiconductor spectrometers were used, since they have higher sensitivity in this energy domain and better energy resolution. These experiments delivered the yields of the discrete photon transition instead of the smoothed energy distributions.

These techniques allowed to measure the γ-rays with energies from ≈ 10 keV up to ≈ 190 MeV.

The technique used to measure the Prompt Fission Gamma Spectra (PFGS) from ^{252}Cf attributing them to the specific fission fragment (FF) mass. Several experiments could be found in the literature where the PFGS were measured in coincidence with fission fragments having certain kinetic energy. Kinetic energy can be converted into the certain fission fragment mass or range (mass split). To get this information the flight time distribution between fission event and fragment detection was measured.

Generally, such mass differential data allow to obtain the aggregate PFGS for ^{252}Cf(s.f.), if the γ-spectra were measured for practically all fragments. Conversion of mass differential spectra in total one requires the fission fragment independent mass yields for ^{252}Cf(s.f.) which are known nowadays with sufficient uncertainties.

Such measurements are relative seldom due to their complexity and, moreover, they usually provided gamma spectra in the narrow energy range and without absolute normalization.

The technique used to measure the Total Fission Gamma Spectrum (TFGS) from ^{252}Cf. The main difference of the total gamma spectra measurements is the absence of the fission event detection and γ-f time selection. All γ-rays accompanying the fission of ^{252}Cf at any emission time are registered. These experiments usually used the strongest and an encapsulated Cf sources. Isotope ^{252}Cf was distributed in the matrix surrounded by one or two capsules which provide the robust and hermetic storage of radioactive substance. The capsules walls have a typical thickness of ≈ 1 mm and were made of aluminium, copper, iron or of alloys on their basis. The activity of the encapsulated Cf source is typically within 400 – 7200 µCi, thus exceeding the Cf source strength used for prompt γ-ray measurements by one or more orders of magnitude, see Table 4.1.

Average Multiplicities and Energy of prompt and total γ-ray emission from ^{252}Cf(s.f.). Experiments, in which the prompt or total γ-ray energy spectra were measured, often provide the average γ-ray multiplicity M_γ or spectrum average γ-ray energy ε_γ. The authors usually obtained these quantities from the integration of the measured γ-ray energy distribution. In several cases, when the average energy was not reported, we calculated it ourselves employing the original energy spectrum.

Besides the integral quantities derived by authors from the measured γ-ray energy spectra, several works measured such characteristics directly or reported them without providing the gamma energy spectra.

4.2. Measurements of the prompt fission γ-ray spectrum

This sub-Section collects and overviews all known experiments where the prompt fission gamma spectra (PFGS) or prompt γ-ray average multiplicities and energies were measured. The measurements are presented in chronological order starting from the first relevant publications. The overview summarizes information important for evaluation: the key details of experimental set-ups, the PFGS measuring technique, primary data processing and applied corrections, presentation of data in publications, availability of uncertainties, etc.

Table 4.1 lists found measurements of PFGS and Table 4.2 – average prompt γ-ray multiplicity Mγ(p) and average energy Eγ(p) per spontaneous fission. Values in the latter were obtained by experimenters who integrated the measured prompt energy γ-ray spectrum. Practically all numerical experimental data were taken from EXFOR [3] (several original data sets not available in EXFOR were compiled during this work).

The top parts of Figs 4.1, 4.2 and Fig. 4.3 plot the prompt γ-ray energy distribution reported in the measurements listed in Table 4.1. Since the PFGS changes by six orders of magnitude the plots represent the three γ-ray energy intervals: 0.08 to 1.0 MeV (where the resonances or oscillations are definitely visible), 1.0 – 10.0 MeV (smooth and exponentially decreasing part of spectra) and 7 – 200 MeV (high energy tail exponentially decreasing up to energy ≈ 20 MeV, then not definitely defined by strong scattered experimental data). To demonstrate the quantitative (dis)agreement of measured data in a reasonable scale the bottom parts of Figs 4.1 and 4.2 display the ratio of measured spectra to the analytical function which is a weighted sum of two Maxwellian distributions. It was difficult to find an optimal analytical representation of the PFGS in the wide range from 0.1 to 20 MeV, thus the ratio still oscillates around unity.

The original experimental gamma spectra are usually normalized per one spontaneous fission (f) event. There are a few exceptional cases where the data were presented by authors in arbitrary units. The prompt γ-ray energy spectra data (measured in coincidence with fission events) are denoted in Figs 4.1 – 4.3 by the open symbols, the total γ-ray spectra (measured without detection of the fission events) by closed symbols.

In total, 19 experiments were carried out to measure the energy distribution of γ-rays accompanying the spontaneous fission of ^{252}Cf in various laboratories in the period from 1956 to 2022. In the following, they are surveyed and analysed from the point of view of inclusion in the GMA fit.

(1) In 1956, A. Smith and colleagues were the first who measured the prompt γ-ray energy spectra from the spontaneous fission of ^{252}Cf [18]. The recently developed scintillation gas counter [19] was used to accommodate the Cf-source and to register prompt fission events by photomultiplier. It served as an excellent fast-fission detector, combining good energy resolution with high speed and an insensitivity to gamma radiation. The gas cell was coated with carrier-free ^{252}Cf yielding about 5000 fissions/minute.

Two methods were utilized for the γ-ray spectrometry. In the first the pulse-height distribution from a single 1 × 1½ inches NaI(Tl) crystal is measured in 0.3 µs time coincidence with the pulses from the fission detector. In the second approach, a double-crystal Compton spectrometer was used in ≥ 2.5 µs coincidence with the fission detector. The former method was the most sensitive method available, the latter gave a unique response to the incident quanta due to selection of Compton backscattered ones. By combining the results of the single and double-crystal measurements, the composite spectrum of prompt fission γ-rays was obtained.

The incident photon spectrum from the NaI(Tl) detector pulse-height distribution was determined with an iterative method. The energy calibration and absolute normalization were checked against the calibrated sources.

A. Smith et al. also reported the prompt γ-ray multiplicity 10.3 γ/f, however without uncertainties [18]. *Unfortunately, the authors neither discussed nor provided any uncertainties of measured data. We derived information about the energy resolution of the NaI(Tl) spectrometer from the spectrum of two "half-resolved" γ-lines of ^{60}Co (Eγ = 1.173 and 1.333 MeV), plotted in one of the figures in [18]. The relative energy resolution was then estimated as ΔEγ ≈ 0.5 × (1.333 - 1.173) / (1.333 + 1.173) /2 ≈ 6.4%. Regarding the year of publication and absence of several experiment details we supposed ± 10% systematic and ± 20% statistical uncertainties for the γ-ray spectrum measured by A. Smith et al.*

(2) 1n 1958, H. Bowman and S. Thompson measured the multiplicities and energy distribution of the γ radiation in the time coincidence with single-fission events [20]. The tiny amount of ^{252}Cf was deposited on the 5 mg/cm^2 plastic film and was placed between the two halves of a two-sided Frisch-gridded ionization detector. The ionization produced by the two fission fragments was measured and recorded in time coincidence. The fission fragment detectors were calibrated by replacing the ^{252}Cf source with ^{235}U and by exposure to a thermal neutron flux. The pulse-height spectrum of gammas was measured with a NaI(Tl) spectrometer. Care was taken to correct for or to eliminate the simultaneous detection of two or more γ-rays per fission event. The resulting γ-ray spectrum was obtained from the apparatus spectrum employing the response function of the NaI(Tl) detector. The average multiplicity was reported to be 10 and the average total energy 9 MeV [20].

No more information relevant for the present analysis was given by the experimenters in this single and short publication [20]. The ^{252}Cf(s.f.) prompt γ-ray energy spectrum is presented there only as a plot of the number of events per fission event versus the γ-ray energy Eγ. We supposed that the plotted data are an integral N(E) of γ-ray spectrum S(E) after integration from the low limit E until the maximum possible energy: $N(E) = \int_E^\infty S(E_\gamma)\, dE_\gamma$. *Then we derived the prompt energy spectrum as* $S(E) = -(N(E_{i+1}) - N(E_i))/(E_{i+1} - E_i)$. *It is plotted in Fig. 4.1 after multiplication by 1000. As can be seen, the Bowman data, after described processing, reasonable agree with the others. For Eγ > 1.1 MeV the large systematic and increasing underestimation is observed – these points were excluded from the GMA analysis. The statistical uncertainty ± 20% was assigned to the used points of this rather old experiment. The spectrum was considered as having no absolute normalization, i.e. as a shape.*

(3) In 1969, N. Ajitanand measured the prompt γ-rays (and neutrons) spectra by NaI(Tl) spectrometer in coincidence with fission fragments [21]. He used the "weightless" source of ^{252}Cf (spontaneous fission rate of 3 10^5 min^{-1}) deposited over an area of 0.2 cm diameter on a 61.4 mg/cm^2 thick platinum backing. The fission fragments (FF) were detected by a silicon surface-barrier semiconductor positioned at a distance of 1.1 cm from the source. Both the Cf-source and FF-detector were installed in the evacuated chamber. The 5 cm × 5 cm NaI(Tl) crystal was used to detect the gamma rays at a distance of 6.5 cm from Cf source. The fission γ-rays (and neutrons) detected by the NaI(Tl) crystal had to pass through the platinum backing of the Cf source foil and a 0.025 cm thick brass window of the chamber.

The amplified pulses from the detectors were fed directly to a coincidence unit which had a resolving time of 1 µsec. The usual calibration procedures were employed for calibrating both detection channels and they were repeated frequently between experimental runs to check the overall stability of the electronic instruments. After subtraction of chance coincidences, the results were corrected for contributions from fission neutrons (8% for the binary fission). The error introduced in the calculated effective solid angles due to errors of geometrical measurement was 2%.

The measured γ-ray spectrum is presented in this single paper [21] as a plot of counts per fission versus the NaI(Tl) pulse height channel. Fortunately, the second scale for Eγ was also given in that plot. It allowed us to convert pulse height distribution into a gamma energy spectrum. The resultant data are plotted in top part of Fig. 4.1 and confirm that the Ajitanand' measurements were not absolutely normalized, consequently we used them in GMA fit as a "shape". The data from [21] were digitized and compiled in EXFOR with the following comment "due to poor quality of the figure, the digitized value of six data points above gamma energy of 1463.56 keV may not be good". This is a reason why we use the N. Ajitanand' data in our evaluation up to energy 1.399 MeV. We added ± 20% statistical uncertainty to every accepted point.

(4) In 1973, V. Verbinski et al. published the results of their measurements of the spectra of the prompt γ-rays from ^{252}Cf(s.f.) emitted within 10 ns after fission [22]. A γ-ray spectrometer consisting of a NaI crystal of 5.85 cm diameter and 15.2 cm length was surrounded by a NaI anti-Compton sheath and had a near-Gaussian response to gammas. The spectrometer was placed 70 cm from the Cf-source that allowed the excellent time of flight separation from the prompt fission neutrons. The background due to chance coincidence events was measured and subsequently subtracted.

The fission fragments were detected by the Si surface barrier detector, which provided a zero-time reference for fission events. The detector was located at 0.25 cm from the foil with deposited ^{252}Cf. The short distance provided the wide acceptance angle of the fission fragments. The non-isotropic angular distribution of γ-rays with respect to the direction of the fission-fragment trajectories was shown by calculation to be negligible for this experiment.

The authors of [22] converted the measured pulse-height distribution into a γ-ray spectrum by using a dedicated spectrum unfolding code. An input for this operation, a response matrix for monoenergetic γ-rays, was measured with several γ-ray sources and then fitted with analytic functions.

The measured γ-ray spectrum is presented in paper [22] in form of a plot (Fig. 9) and for a number of energy intervals in tabulated form. The error bars shown in the plot only represent the statistical errors which are mostly due to the efficiency calibration of the γ-ray detector. There were also systematic errors which are mostly due to the NaI detector efficiency calibration. We derived the latter from bars in Fig. 5 "Absolute photopeak efficiency of the NaI spectrometer" in [22]: the systematic uncertainty increases from ± 5% at γ-ray energy 0.14 MeV to ± 20% at ≈ 7 MeV.

(5) F. Dietrich with colleagues measured the spectrum of the prompt γ-rays for ^{252}Cf spontaneous fission in the 8 to 20 MeV range [23]. The ^{252}Cf source of 3.7 10^{+4} f/s was evaporated onto a 5 cm^2 nickel foil and placed in an ionization chamber, the signal from which served for counting and timing.

The γ detector was a 25.4 cm diameter × 25.4 cm long NaI scintillator surrounded by a 15.24 cm thick NE102 plastic anticoincidence shield on the sides and a 3.81 cm thick shield in front. This shield reduced cosmic-ray background and improved the energy resolution of the system. The γ-spectrometer was additionally protected by surrounding layers of Pb and ^{6}LiH. The distance to the Cf source was either 43 cm (where the solid angle subtended by the detector was 0.251 sr) or 89 cm (where two solid angles 0.0179 sr and 0.0365 sr were arranged with the help of two different collimators).

The experimenters clearly separated prompt γ events by TOF from the Cf fission neutrons and those have undergone multiple scatterings on the way to the NaI crystal. The full width at half-maximum (FWHM) of the γ-ray peak was 2.4 ns, which allowed the selection of prompt gamma within 20 ns time window.

The energy calibration was accomplished by using a Pu-Be neutron source, which provides γ peaks of 2.23 MeV from neutron capture in the plastic shield, 4.43 MeV gammas from $^{9}Be(\alpha,n)^{12}C^{*}$, and 6.797 MeV from full-energy capture of slow neutrons on ^{127}I. The Livermore tandem accelerator was used to generate monoenergetic line shapes for calibration of the spectrometer for the different geometries used in this experiment. Thus, the γ rays of energy 14.0 and 16.8 MeV were produced by the $^{15}N(p,\gamma)^{16}O$ reaction at incident proton energies 2 and 5 MeV, 4.43 MeV by the $^{15}N(p,\alpha)^{12}C^{*}$ reaction. To determine the absolute yield, the accepted γ pulse-height spectra were unfolded by an iterative technique using response functions for discrete γ lines measured with the identical geometries used in the ^{252}Cf source runs.

The experimenters reported the following information about uncertainties and corrections [23]. In the low-energy domain the extrapolation of the line shapes contributed an uncertainty of ± 15% to the final absolute scale. The counting statistics in the original spectra were amplified by the unfolding procedure; these variations were reduced by Gaussian smoothing with FWHM in the range 2 - 4%. The absolute yield was corrected for a 12% absorption of γ rays in the front ^{6}LiH shield, the front plastic and the NaI housing materials, and for the γ rays that passed through the NaI without interaction (calculated as 3% for the two small solid angles).

We used the γ-ray spectra measured by F. Dietrich et al. [23] as three sets of data with absolute normalization uncertainty ± 15% and statistical bars obtained during digitizing of the plot. We assigned ± 15% to the latter. The measurements at 89 cm distance (solid angles 0.0179 sr and 0.0365 sr) were considered to be intercorrelated with factor 0.5. The γ-ray spectrum obtained at 43 cm with the solid angle 0.251 sr was supposed to be cross correlated with two spectra measured at a distance of 89 cm by factor 0.3.

(6) In 1989, J. Kasagi et al. measured the spectrum of the prompt γ-rays from ^{252}Cf(s.f.) with energies from 5 to 155 MeV [25]. It was the first experimental investigation of the γ-ray yield beyond 20 MeV showing a substantial change of the slope of the spectral shape. Authors attributed these γ-rays to the coherent nucleus-nucleus bremsstrahlung emission. At gamma energies lower than 20 MeV their data are in good agreement with previously reported data confirming the statistical decay mechanism of the excited fission fragments.

The ^{252}Cf source of 3.9 10^{+4} f/s was contained in a standard capsule. It was placed next to a NE-213 detector of 12.5 cm diameter and 5 cm length, which detected γ-rays emitted from fission fragments and provided a zero-time reference. The neutron induced events in the NE-213 detector were almost completely rejected by pulse shape discrimination. The detector threshold was set just above noise level which resulted in a 36% detection efficiency of the total fission events.

The γ-ray spectra were measured by the BaF_2 which consisted of seven optically separated hexagonal crystals, each 30 cm^2 × 20 cm. By registering and combining the optical signals from all crystals, the total energy deposition of γ-rays was eventually measured. A narrow gate of 2.9 ns set around the prompt γ-peak in the time-of-flight spectrum served to reject the events induced by neutrons and to reduce the random coincidence events at a level below 0.3%, which were subtracted from the spectrum.

The background was measured without the ^{252}Cf source. Only energetic cosmic rays can contribute to the background and may be observed in the coincidence measurements. The authors noted that the time spectrum of the background run had a sharp peak at the same position as in the run with the Cf source. They explained this with the background events caused by the bremsstrahlung induced by the cosmic high energy charged particles in the NE-213 detector. Other sources of the spurious events (pulse pile-up, (n,γ) reactions on source nearby materials) were investigated, but they proved not to affect the high energy spectrum.

The measured γ-ray spectrum is presented in short publication [25] as a figure, from where the data were digitized for EXFOR. We used these numerical data in the GMA fit as an absolute ^{252}Cf(s.f.) γ-ray spectrum. The digitized uncertainties which range from 18% at 16 MeV to ≈ 100% at 155 MeV were considered as statistical (the similar uncertainty 18% was also assigned at Eγ < 16 MeV). Since the efficiency of the fission events registration by the NE-213 detector was essentially below 100% and since its uncertainty was not specified by the authors, we assigned a ± 10% systematic uncertainty to the Kasagi' data.

(7) In 1990, Yu.N. Pokotilovskii measured the emission of high energy γ-rays in the energy range 20 – 160 MeV [26] to check the experimental data of J. Kasagi et al. [25]. He used a ^{252}Cf source with a strength of about 7 10^6 fission per second. A big volume NaI(Tl) detector of 15 cm in diameter and 15 cm in length was placed at a distance of 38 cm from the source. In front of it a lead collimator was positioned between source and detector which restricted the illumination of the NaI(Tl) detector within a circle of Ø6 cm. Additionally, a 25 cm long absorber of borated polyethylene was used to reduce the flux of neutrons and γ-rays with an energy below 1 MeV while weekly impacting on the energetic gammas. The NaI(Tl) scintillator was surrounded by a system of 2 cm thick flat plastic scintillation counters which worked in anticoincidence. This decreased the cosmic rays background by several decades. The measuring procedure consisted of runs with and without a 1.5 cm lead absorber, which reduced the γ radiation in the energy range of interest by several times.

The energy calibration of the spectrometer was accomplished with a set of discrete γ-lines from the calibrated radioactive γ-sources and by 4.4 MeV gammas from the reaction ^{9}Be(α,n)^{12}C*. An additional calibration was carried out by means of cosmic rays.

The results of the Yu. Pokotilovskii measurements [26] are presented as the upper limit of the γ-ray spectrum in the energy range 20 – 140 MeV. This upper limit is lower than the results of J. Kasagi et al. [25] by more than an order of magnitude.

The data of Yu. Pokotilovskii [26] were not used in the present GMA evaluation, since only the upper limit for the ^{252}Cf(s.f.) γ-ray spectrum was given.

(8) In 1991, E.A. Sokol et al. measured the ^{252}Cf(s.f.) γ-ray spectra from spontaneous fission of ^{252}Cf, ^{254}Cf and other heavy nuclei [27], [28]. To detect the γ-rays, an assembly of eight scintillation detectors based on bismuth-germanate ($Bi_4Ge_3O_{12}$) crystals was employed. Each crystal had a diameter of 3.6 cm and length of 14 cm and was packed in aluminium case. A vacuumed steel cylinder with two Si semiconductor detectors and a “point” Cf source were positioned in the centre of the assembly.

For any fission event, the pulse heights (PH) from each of the eight scintillation detectors were collected during 4 µs after detection of fragments. The PH spectra of one, two, ... up to eight γ photons, and the total spectrum, regardless of the number of photons, were detected in the energy range 0.320 – 5.120 MeV. The measured pulse height distributions were converted into γ-ray energy spectra applying the method of statistical regulation. The response functions of the scintillator detector assembly were determined experimentally with the help of γ-lines with energies 0.898, 1.115 and 1.836 MeV from calibrated γ-sources and 4.43 MeV from the Pu-Be source (^{9}Be(α,n)^{12}C* reaction). The response functions were also calculated by the Monte Carlo method.

The authors of [28] observed that the amplitude distributions were similar for all the nuclei they studied. Therefore, they determined the absolute characteristics of the γ radiation of ^{252}Cf, ^{254}Cf and other nuclei using the absolute values for ^{252}Cf of V. Verbinsky et al. [22]. The necessary corrections for different neutron counting and recording threshold 320 keV were applied by experimenters. The uncertainties for the integrated values (gamma multiplicities and average energies) amounted to 5 - 7%.

The ^{252}Cf(s.f.) γ-ray spectrum of E. Sokol et al. [27], [28] was used as the relative (shape) data set. As can be seen in Fig.4.1, the measured spectrum starts systematically underestimate the bulk of other data for Eγ < 1.0 MeV and overestimate for Eγ > 4.1 MeV. Consequently, the low and high energy parts of his spectrum were excluded. The statistical uncertainty of ± 20% was assigned to all points of this experiment, since no relevant information was reported by the authors.

(9) S.J. Luke et al. tried to measure the prompt γ-rays spectrum from ^{252}Cf(s.f.) in the energy range from 5 MeV to ≈ 130 MeV [29], [30]. The team used two different techniques for the detection of high energy γ-rays: direct coincidence with fission fragments (FF) and coincidence with any prompt γ ray. The second method was used to reproduce, as closely as possible, the experiment of J. Kasagi et al. [25].

The γ-FF coincidence measurement used a 50 µCi ^{252}Cf source deposited on a platinum disk. The Cf source was placed in the centre of a scattering chamber at a distance of 1.2 cm from a silicon surface barrier detector. The silicon detector was collimated to yield a half angle to 22° and 38°, depending on the size of the detector. A 25.4 cm × 38.1 cm NaI detector was 50 cm away from the Cf source and was surrounded by a passive lead shield and an active anticoincidence shield to minimize the cosmic-ray background. The solid-state detector was placed at an angle of 120° with respect to the axis of the NaI detector to maximize, as much as possible, the detection of higher multipole radiation, particularly the quadrupole radiation. In addition, the time of flight between events in the fission-fragment detector and in the NaI detector was recorded. The gate in the time-difference spectrum was narrow enough to allow the discrimination of neutrons with energy 25 MeV or less.

The γ-γ coincidence measurement was performed by placing a NE-213 liquid scintillator 25.4 cm from the Cf source resulting in a 20% geometric efficiency for the detection of prompt γ-rays. Time-of-flight and pulse-shape discrimination were used to separate neutrons from photons in the scintillator. This discrimination allowed to separate photons from neutrons with energies lower than 44 MeV. The liquid scintillator was placed collinear with the axis of the NaI detector. The angular correlations between the fission fragments and the high energy γ-rays were not important, since the prompt γ-rays are emitted roughly isotropically from the fissioning system.

The NaI detector was calibrated up to a γ-ray energy of 30 MeV using low energy sources and γ-rays from reaction $^{11}B(p,\gamma)$ at E_p = 7.25 and 14.3 MeV. The calibration was also checked against the 123 MeV cosmic-ray muon peak in the NaI spectra, which reproduced the energy of this peak to better than one percent.

The experiment of S. Luke et al. [30] could not reproduce the high energy gamma emission rate from ^{252}Cf(s.f.) at the level reported by J. Kasagi et al. [25], neither in the γ-FF nor γ-γ modes.

The γ-ray spectrum measured with the fission-fragment and γ-ray coincidence mode in the medium energy range energy, 5.3 MeV < Eγ < 21 MeV, is presented as a figure in [30], from where the data were digitized for EXFOR. The uncertainties given in the plot, 12 – 94%, were considered to be statistical. An additional systematic error of ± 10% was assumed for the geometry efficiency of the Si detector located at some distance from the Cf source.

(10) In 1992, H. van der Ploeg et al. measured the γ-ray energy spectrum and the angular dependence relative to the fission direction of photons in the energy range 2 to 40 MeV for the spontaneous fission of ^{252}Cf [31].

A californium source of 1.2 10^4 fission decays/s was used during 26 days of effective data taking. Four low-pressure position-sensitive avalanche detectors (PSAD) were placed around the Cf source within the vacuum chamber. They were oriented in such a way that, for each high-energy gamma event detection, the fission fragment could be observed either along or perpendicular to the direction of the γ-ray emitted. One ninth of all fission decays were detected.

Two 2.6 litre volume BaF_2 detectors and a 25 cm diameter × 35 cm cylindrical NaI spectrometer with a plastic anticoincidence shield were used to detect high-energy γ-rays in the range of 2 to 40 MeV. Eight 3.5 × 3.5 × 6.0 cm^3 BaF_2 gamma detectors, four below and four above the reaction plane, were located close to the ^{252}Cf source and were used to determine the start signal for each event. Time of-flight measurements distinguish prompt γ-rays from neutrons for the large-volume gamma detectors. The γ-ray background, due to cosmic radiation, was efficiently suppressed by measuring coincidences with the fission fragments. For the NaI detector, further reduction is obtained by plastic anticoincidence shielding.

A strong anisotropy, W(90°) / W(0°) ≤ 1.5, of γ-rays with energies 8 MeV < Eγ < 12 MeV was observed when they have a direction perpendicular and parallel to fission fragments detected by PSADs. Based on this observation, the authors [31] suggested that in the fissioning system γ radiation may occur prior to nuclear scission.

The measured prompt gamma spectrum for ^{252}Cf(s.f.) in the energy range 2 to 37 MeV is presented in a Figure in [31], from where the data were compiled in EXFOR. Statistical uncertainties comprise: 17 – 100% are the bars digitized for E_γ = 13 - 37 MeV and 10% as extrapolation or our assumption for E_γ < 13 MeV. A systematic error of ± 10% was assigned to these data since no error analysis is given by authors.

(11) D. Pandit et al. measured the energy spectrum of the prompt high energy γ-rays in coincidence with the prompt low energy discrete γ-rays emitted from the decay of excited fission fragments of the spontaneous fission of ^{252}Cf [32].

The 3 μCi ^{252}Cf source was placed as close as possible to the four BaF_2 detectors, arranged in a 2 × 2 matrix, to get a start trigger in order to separate and reject the neutrons and cosmic rays.

The high energy γ -rays were measured using the array of 49 BaF_2 crystals assembled in a 7 × 7 matrix at a distance of 35 cm from the ^{252}Cf source [32]. A master trigger was generated by taking a coincidence between the start trigger and any one of the 49 detectors in the pack above the 4 MeV threshold ensuring the selection of fission events and rejection of background. The time of flight measurement distinguished the gamma-rays from neutrons while long/short gate technique was applied to reject the pile up events.

At the photon energies Eγ ≥ 25 MeV, cosmic ray showers were the major source of background. To suppress the background lead bricks were used as a passive protection shield by experimenters. Large area plastic scintillator pads were used as active shielding that surrounded the BaF_2 array as well as the multiplicity filter to reject the cosmic muons. In addition, the cosmic pile up events were rejected using the cluster summing technique developed by the authors. Finally, the random coincidence events were subtracted measuring the background spectrum without the Cf source in an identical configuration [32].

According to the information given in the EXFOR entry 33041, the measured γ-ray spectrum data were received from the authors of this experiment [32]. The data set represents the absolute ^{252}Cf(s.f.) prompt γ-ray spectrum from energy 4 to 79 MeV with statistical uncertainty varying from about 0 to 95%. We assigned 100% at several high energy points where also a zero value for the γ-spectrum was reported. The systematic uncertainty is not given in [32]. Neither was there information on the γ-ray detector calibration and conversion of the pulse distribution into the gamma energy spectrum. Due to this an additional ± 10% uncertainty was assigned to the data set of D. Pandit et al.

(12) In 2011, N.V. Eremin et al. measured the probability of high-energy γ-ray emission accompanying the spontaneous fission of ^{252}Cf nuclei in the energy range of 4.4 – 60 MeV [33] - [35]. The technique of γ-quantum coincidence with fission neutrons in combination with the time of flight method was used in this work.

The experimental set-up included a pair of scintillation detectors based on bismuth trigermanate (a BGO detector of γ rays) and polystyrene (a plastic detector of neutrons). The BGO crystal with a size of Ø7.6 × 7.6 cm and the polystyrene organic scintillator with a size of Ø6.0 × 2.0 cm were connected to photomultipliers and a digital storage oscilloscope. BGO detectors were used to measure the γ-ray energy. The plastic scintillator detector was able to detect γ-rays as well as neutrons. The axes of the two BGO detectors in respect to the axis of the plastic detector formed the angles 90^o and 180^o, respectively.

A ^{252}Cf source had an activity of 6.1 10^6 neutrons per second [33] - [35]. The distance between the source and BGO detector was set at 10 cm so that the probability of simultaneous detection of two γ quanta was lower than that of detection of one γ quantum. The distance between the source and the plastic detector was chosen at 50 cm to distinguish neutrons from photons by the time-of-flight (TOF). The signals from the plastic detector served as a ”start”, from BGO detectors as a ”stop”. The time resolution derived from the γ-γ coincidence peak in the TOF spectrum was 5 ns. The main time window with width ≈ 20 ns selected the true neutron-gamma coincidences. Additionally, another two windows were set to estimate the random coincident events.

The experimenters calibrated absolute efficiency for the γ-ray detecting system with the help of the transport simulation code GEANT4. The source of γ-rays was modelled in the geometry of the experimental setup. For γ-rays of 70 MeV, depositing 65 – 70 MeV of its energy in the BGO, the absolute detecting efficiency was about 0.2%. Standard γ-sources ^{137}Cs, ^{60}Co and Pu-Be were used for the energy calibration of the BGO detector. At higher energies, the gamma detector efficiency was determined by measuring the energy losses of cosmic muons in the BGO crystal in coincidences with the plastic detector.

The data processing included the rejection of pile-up events to be sure that the γ-ray energy was measured accurately [33]-[35]. Digitization of the shape of pulses from detectors and their digital processing allowed to reject overlapping. The influence of cosmic γ-ray events was measured in the same geometry but without Cf source.

The experimental data with uncertainties of N. Eremin et al. were presented as a plot in their publications [33]-[35], from where they were digitized for EXFOR - we used them in the present GMA fit. The uncertainty components were not discussed in Refs. [33]-[35]. As statistical ones we used those given in EXFOR 41551003: around 40 – 100% for Eγ = 14 – 60 MeV, then assigning 30% below 14 MeV. In addition, a 20% systematic uncertainty was supposed to account for all possible uncertainties not quantified by the experimenters.

(13) In 2012, E. Kwan and colleagues measured the energy distribution of prompt γ-rays from the spontaneous fission of ^{252}Cf and neutron-induced fission of ^{235}U at the WNR facility of the Los Alamos Neutron Science Center (LANSCE) using the FIGARO neutron detector array [36].

The array held 17 EJ301 organic liquid scintillators, each with active volumes of 12.5 cm in diameter and 5.0 cm in depth. The detectors were positioned ≈ 1 m away from the source covering angles from 42^o to 125^o. Out of 17, only six detectors with the best pulse-height distributions were chosen for the analysis. The particles were identified using the double time-of-flight technique, which measured the time difference between the source pulse and the fission event as well as between the fission event and the neutron detectors.

Fission events were detected by the Parallel Plate Avalanche Counter (PPAC), which was designed to minimize the amount of structural material that can scatter neutrons [36]. A counter for the measurement with Cf contained a single foil placed at the center of the fission detector. A deposit of ≈ 2 μCi ^{252}Cf was on both sides of a 4 cm in diameter and 0.3 cm thick titanium backing foil. Two 1.4-mm thick double-sided aluminized mylar foils sandwiched the target and served as the cathode for PPAC. The anodes positioned 3 mm away from the cathode consisted of the same type of aluminized mylar foils.

The prompt γ rays concentrated at around 3 ns in the time-of-flight spectrum were clearly resolved from the prompt neutrons that occur more than 20 ns later. The fission gamma rays were also differentiated from fission neutrons by using the fast and slow components of the scintillation pulse.

The experimenters simulated the response of the liquid scintillators as a function of incident γ-ray energy by a GEANT4 code with the built-in geometric model of the detector array including the PPAC and its environment. The calculated detector responses were validated against the measurements with a Pu-Be mixed neutron and γ-ray source and four standard calibration sources ^{22}Na, ^{88}Y, ^{60}Co and ^{137}Cs.

The authors of [36] chose the single value decomposition (SVD) and the iterative Bayesian methods to deduce the incident γ-ray distributions. Both methods predicted exponential decays with similar slopes above 1 MeV, but disagree below 1 MeV. The Bayesian method indicated that there is a broad peak in the distribution around 300 keV, while the SVD technique suggested a much broader and smoother curve.

For EXFOR entry 14413005, E. Kwan and colleagues [36] provided the numerical data for the ^{252}Cf(s.f.) prompt γ-ray spectrum obtained by the Bayesian unfolding method in arbitrary units. This spectrum, after our re-normalization to the bulk of other data, is displayed in Fig. 4.1. The visual comparison indicates that for Eγ ≤ 0.36 MeV the spectrum of E. Kwan et al. falls to zero unreasonably quickly. Regrettably, but the authors provided no uncertainties. The single information available in their publication [36] is a ~ 0.3% statistical uncertainty for the response matrix. We used the data of E. Kwan et al. from entry 14413005 as arbitrary prompt γ-ray spectrum above 0.44 MeV. The statistical uncertainties were taken from the Fig. 6 of [36] where they are given for Eγ > 4.12 MeV, then we assigned ± 10% for all other points below.

(14) 2012 to 2014, A. Chyzh and colleagues published the prompt γ-ray energy distributions for the spontaneous fission of ^{252}Cf measured at the Los Alamos Neutron Science Center (LANSCE) [37], [38], [39]. The prompt γ rays emitted in fission were detected by the Detector for Advanced Neutron Capture Experiments (DANCE) in coincidence with the detection of fission fragments by a Parallel-Plate Avalanche Counter (PPAC). In [39] the same data set is analysed as in [37] and [38], but with a different technique and it additionally reports the total prompt γ-ray energy distributions, obtained by the unfolding of the measured two-dimensional spectrum of total γ-ray energy versus multiplicity.

A ^{252}Cf source with a strength ~0.15 μCi was deposited on a 3 μm thick titanium foil and covered by a 1.4 μm thick aluminized mylar to serve as a cathode. The two anodes, made of the same thickness aluminized mylar sheet, were placed on both sides of the cathode at a distance of 3 mm and electrically connected. The PPAC was operated with isobutane gas at ~4.00 torr pressure. It provided a fast fission trigger for the DANCE array to record the coincident γ rays from fission fragments within a time window of ~ 30 ns.

The DANCE spectrometer is a 4π highly segmented and efficient γ-ray calorimeter. It consists of 160 BaF_2 scintillation crystals with a length of 15 cm [38]. The crystals formed the sphere with the inner radius of 17 cm. The hardware threshold for detecting γ-ray energy by DANCE was 150 keV. The detection efficiency varies between 85 and 88% for γ-ray energy between threshold and 10 MeV. The efficiency and response matrices were derived from the simulations by GEANT4 with a geometric model of DANCE and surrounding materials. The model was validated by comparison with the measurement of γ-ray calibration sources ^{22}Na, ^{60}Co, and ^{88}Y.

The experimenters also reported in [37] that the average γ-ray multiplicities are 8.16 and 8.14 γ/f derived by the Bayesian and SVD unfolding methods, respectively (i.e. a consistency better than 1% was achieved between these two methods). The spectrum averaged γ-ray energy ε_γ from both Bayesian and SVD unfolded spectra are 0.94 and 0.98 MeV, respectively, with the accuracy better than 5%. *From this we derived the average total γ-ray energy* $E\gamma = \varepsilon_\gamma \times M\gamma = <0.94 + 0.98\ \mathrm{MeV}> \times <8.16 + 8.14\ \gamma/\mathrm{f}> = 7.824\ \mathrm{MeV/f}$.

The prompt γ-ray energy spectra are compiled in EXFOR entry 14143.002 (two sets of data obtained by unfolding either with the iterative Bayesian or SVD methods) and in 14361.011 (one set of data obtained using by Bayesian deconvolution method). All data are provided in numerical form by the authors of the experiment and have the arbitrary normalization. We used two sets in GMA: one from 14143.002 (unfolding with SVD method) and one from 14361.011 (unfolding by Bayesian method). Since the difference between them is a deconvolution method of the same measured pulse height distribution, we suppose a 50% correlation among ± 10% systematic uncertainties (that reflects equal contribution of the measured and unfolding components to the uncertainty).

(15) In 2015, A. Oberstedt et al. reported the results of six measurements of the fission prompt γ-ray spectra which involved five different lanthanide halide detectors at the Joint Research Centre (IRMM) [40], [41].

The author's most recent publication [41] reported new measurements with a ^{252}Cf source located inside a cylindrical vacuum chamber. An artificial polycrystalline chemical vapor deposited (CVD) diamond detector with an active area of 1 cm × 1 cm, which was mounted directly above the ^{252}Cf sample, provided the fission event fast trigger signals. A coaxial cerium-doped lanthanum chloride ($LaCl_3$:Ce) scintillation detector of size 1.5 × 1.5 inches (labelled SEB 347) was placed at a distance of 37 cm from the fission source in order to measure photons in coincidence with fission fragments (data are compiled in EXFOR entry 23197.010). A similar set-up was used in other measurements, but with coaxial cerium doped lanthanum bromide ($LaBr_3$:Ce) scintillation detectors: two detectors of size Ø2.0 × 2.0 inches (labelled Q489 and Q491) at distance 40 cm from Cf source (measured data are compiled in 23197.011 and 23197.012, correspondingly) and of size Ø3.0 × 3.0 inches (labelled 2987) at distance 60 cm (data are compiled in 23197.013) [41]. These detectors have been also used in previous ^{252}Cf(s.f.)γ experiments of this group [40].

All detectors were placed perpendicular to the symmetry axis of the ionization chamber in the same plane as the fission source. The covered solid angle was about 4π/1000, thus the probability of multiple photon events was decreased by a factor of 1/1000. The observed coincidence timing resolution at Eγ = 200 keV was about ≈ 1.5 ns for the $LaBr_3$:Ce and $LaCl_3$:Ce detectors of γ-rays in conjunction with a diamond detector of fission fragments. Fission γ-rays were selected within ± 3 ns relative to the prompt peak in the time-of-flight spectrum. The response functions of the scintillation detectors were determined by simulations with the Monte Carlo code PENELOPE2011 considering all experimental conditions [40]. The measured apparatus energy spectra, after background subtraction, were convoluted into the real emission prompt fission γ-ray spectra.

The experimenters derived the average prompt γ-ray multiplicity Mγ = 8.29 γ/f by calculating the first moment of a measured multiplicity distribution and showed that it is equivalent to an average multiplicity from integrating of a measured PFGS [41].

In the present GMA fit the four prompt γ-ray spectra from $^{252}Cf(s.f.)\gamma$, measured by A. Oberstedt et al. [40], [41] were used. The data were taken from the EXFOR entry 23197, where the numerical data and total uncertainties were submitted by the authors of the experiment. Since the systematic uncertainties are not given explicitly for the energy spectra, we derived them from the uncertainty of the total multiplicity reported by the experimenters for every experimental set-up configuration. The estimated uncertainties of absolute normalization turned out to be rather small 1.5 – 4.0 %. This results to the dominance of the A. Oberstedt' data over all other sets and to the 2% uncertainty of the GMA fit. To avoid this we used a ≈ 5% systematic uncertainty derived from the reported uncertainties for multiplicities Mγ. The statistical uncertainty was derived as square root difference between reported total and found systematic ones. The cross-correlation coefficients between 4 sets of the A. Oberstedt' spectra were supposed to be 0.5.

(16) In 2019, R.P. Vind et al. reported the experimental results of the measurement of the energy spectrum of the gamma rays in coincidence with fission fragments produced in spontaneous fission of ^{252}Cf [42]. The main purpose of this experiment was the measurement of the high energy part of the γ-ray spectrum, namely $E\gamma \geq 20$ MeV.

The fission fragments from a 5μCi ^{252}Cf source were detected by a multi-wire proportional counter (MWPC) of dimension 17.5 × 7.0 cm. It was mounted inside a scattering chamber at a distance 18 cm from the source. The fission events were selected above the threshold energy of the detection of 6 MeV alpha particles emitted from the source. The γ-rays were detected by a Bismuth Germanate (BGO) detector which was 7.6 cm long, had a hexagonal cross section and inner circular diameter of 5 cm. It was mounted at a distance of 1.0 cm from the source. The neutrons were not separated from the gamma rays. The time coincidence information was measured employing the fast signal of the BGO detector as a 'start' and from MWPC as a 'stop' within the time range 200 ns.

The authors of the experiment showed that the gamma yield beyond 20 MeV decreases with increase of its detection threshold [42]. They concluded that these γ-rays are not emitted from ^{252}Cf(s.f.) but can be a contamination due to the cosmic muons piling up with the prompt low energy γ-rays emitted from the fission fragments. According to the authors, further investigation would be required to understand the source of these high energy γ-rays.

The comparison of the γ-ray spectrum measured by R. Vind et al. with other PFGS data showed the systematic different energy shape even below 20 MeV. In [42] no information about the absolute normalization, applied corrections and uncertainties is given. Due to these reasons the data of R. Vind et al. [42] were excluded from the present GMA analysis.

(17) In 2018, L. Qi et al. reported the prompt-fission γ-ray spectra in the range 0.1 to 8.0 MeV from ^{252}Cf(s.f.), measured at the Institut de Physique Nucléaire in Orsay [43].

The fission chamber contained one cathode supporting a 25 mm diameter ^{252}Cf sample [43]. The thin aluminium walls of the chamber minimized scattering of prompt γ-rays and neutrons emitted in fission and thus have minimum distortion of the gamma and neutron spectra. The ionization chamber also possessed the sub-nanosecond time resolution and excellent discrimination between fission fragments and α particles. The number of α particles in the selected fission fragment distribution was estimated to be less than 0.5% and thus induced negligible influence on the measured prompt-fission γ-ray spectrum. PFGS were normalized to the total number of fissions to give average spectral characteristics per fission.

The γ-rays emitted from fission fragments of ^{252}Cf were measured by two types of fast scintillation detectors at typical distance 35 cm [43]: seven individual cerium-doped lanthanum bromide $LaBr_3$(Ce) and a cluster of nine phoswich detectors from the PARIS array. The detectors made of $LaBr_3$(Ce) (5.08 ×5.08 cm and 7.62 × 7.62 cm in diameter and length) had time resolution 300 ps and a good energy resolution 3% at 661 keV. PARIS is an array of a new type of phoswich detectors. The inner shell was $LaBr_3$(Ce) cubic crystals (5.08 × 5.08 × 5.08 cm) and the outer shell consisted of NaI(Tl) rectangular crystals (5.08 × 5.08 × 15.24 cm). The PARIS phoswich detectors benefited from superior energy and time resolution of the $LaBr_3$(Ce) part and increased efficiency, particularly at high energy, from the NaI(Tl) part. The time window was set to 5 ns.

The true emitted PFGS was convoluted with the intrinsic response of the γ detectors using the linear iteration algorithm [43]. The response matrix itself was obtained from detailed GEANT4 simulations of the entire experimental setup. The low-energy threshold was set at 100 keV to exclude the intense X-rays, which cannot be simulated very well. A full energy range up to 10 MeV was used to consider the effect of Compton scattered high-energy γ rays appearing in the lower-energy region of the spectrum. A validation of the response matrix was performed using the detection efficiencies and comparisons between simulated and experimental spectra from conventional γ sources.

The experimenters unfolded the measured spectrum of each individual γ detector separately and the spectral characteristics were then extracted individually [43]. The final values are obtained from the weighted mean and associated uncertainties of spectra from the multiple detectors in the experimental set-up. Each individual unfolded spectrum from multiple detectors was combined into two averaged unfolded spectra for different detector types, i.e., $LaBr_3$(Ce) scintillation detector and PARIS phoswich detector. The authors stated in their paper [43] that both unfolded spectra are in good agreement with each other despite the very different γ-ray responses. There were some small differences observed in the amplitude of the major peaks in the low-energy region, but this may be due to the different energy resolutions of the two detector types and are also within the uncertainties.

The authors also deduced the average multiplicity $M\gamma$ and the total released energy $E\gamma$ by integrating the unfolded spectra. The spectral characteristics for the two types of the detectors agree well within 1.5% and are within the error bars, see Table 4.2.

In the present GMA analysis, we used two prompt γ-ray spectra measured by L. Qi et al. [43] with the help of the $LaBr_3$(Ce) scintillation detector (compiled in EXFOR entry 23417002) and PARIS phoswich detector (EXFOR 23417003). These two sets are considered as absolute energy spectra. The uncertainties are also given in these sub-entries, but no information on their components, neither there nor in [43]. We accepted the 1.8% uncertainty reported by the authors for $M\gamma$ for the experiment with the $LaBr_3$(Ce) and 2.3% with PARIS phoswich detectors as the systematic errors (within which the two data sets are considered to be 50% cross-correlated). An additional systematic 5% error was supposed to account for all other non-quantified components in both data sets.

(18) In 2020. S. De et al. reported the prompt γ-ray spectrum of ^{252}Cf(s.f.) in the energy range 0.1 to 5.14 MeV [44]. The main purpose of this experiment was, however, PFGS from the neutron induced fission of ^{232}Th.

The fission chamber was used to register the fission fragments produced in the ^{252}Cf spontaneous fission. The timing signal from the chamber also served as a trigger for recording the time-of-flight spectrum. Regrettably this publication [44] provides no information about the fission chamber and Cf source.

The prompt fission gamma rays were measured by two 3.81 × 3.81 cm cylindrical $CeBr_3$ detectors, placed at distances of 24 and 26 cm from the ionisation chamber. Lead bricks were used to shield the detectors to reduce the contribution due to background gamma and also to avoid any crosstalk between the two detectors. The gamma ray prompt fission spectra were unfolded by the GRAVEL algorithm which is an iterative technique. The response matrix for the $CeBr_3$ detectors was calculated using the Monte Carlo simulation code GEANT4. This procedure incorporated the parameters obtained in the fitting of the energy spectra of ^{22}Na, ^{60}Co and ^{137}Cs standard γ sources. For incident γ-rays of energies ranging from 0 to 10 MeV in steps of 20 keV, the energy deposited within a bin size of 20 keV was generated [44].

The experimenters compared their results with other measurements available in the literature. They observed that the peak positions obtained in the low-energy region of the ^{252}Cf(s.f.) PFGS agree reasonably well with previous measurements although the energy resolution is inferior as compared to earlier experiments with similar detectors [44].

The authors of [44] estimated the uncertainty involved in the simulation of the response function of the $CeBr_3$ detectors and the inversion technique. It was done by deconvolution of the artificial fission γ-spectrum on the $CeBr_3$ detectors which had gamma peaks of equal intensities. The average percentage deviation in intensities of the unfolded spectrum from the original emission spectrum for energies up to 5.3 MeV was observed to be 3.18%. From comparison with the measurements by R. Billnert et al. [40] and V. Verbinski et al. [22], the authors found that the reproducibility of their measured spectral data was within 3%.

The PFGS data were provided in the EXFOR sub-entry 33135003 by S. De and co-workers in tabulated format. We used them as absolute γ-ray spectrum except the first point at 0.1 MeV which is out of trend. Statistical uncertainties are not given neither in 33135003 nor in [44]. Consequently, we suppose they linearly increase from 10% to 90% to catch the fluctuations of points visible in the Fig. 10 of [44]. Two systematic components are given by the authors [44]: 3.18% resulting from unfolding and 3.0% quantifying the reproducibility from comparison with several others measured PFGS. Such accuracy for the total systematic uncertainty seems too high, this is why we added an additional 5.0%.

In 2018, S. De and co-workers had published the measurements of the prompt fission γ-ray energy spectra in spontaneous fission of ^{252}Cf [45]. It looks like this experiment had a configuration different from the set-up described two years later in [44]. In 2018 they used a Ø7.62 × 15.24 cm inches $LaBr_3(Ce)$ detector at 18 cm from the source. Californium was inside a small ionization chamber operating in air and counting the fission fragments within a 2π solid angle. The ^{252}Cf was deposited on an anode having 4.5 cm diameter, the fission signal was generated in the cathode of the same diameter. A threshold value was set to cut-off the alpha particles. The timing information from the fission chamber and $LaBr_3(Ce)$ detector were used to set a 50 ns window for selecting prompt gammas. The energy resolution of the Lanthanum-bromide detector was found to be 6.94% at 0.34 MeV, 5.00% at 0.66 MeV, 5.797% at 1.41 MeV and 2.98% at 4.44 MeV. The experiment was performed once with the Cf source and then again with the same source but heavily shielded so that no direct γ-rays were able to reach the $LaBr_3$ detector. Then the background was subtracted from the original spectrum.

Unfolding of the measured γ-ray energy spectra has been carried out using GRAVEL method. The response matrix of the detector has been simulated using GEANT4 and the unfolding of γ-ray energy spectra for ^{60}Co and ^{137}Cs sources have been validated [45].

The authors have presented their data in plots and compared with those of V. Verbinski et al. and R. Billnert et al. Similar fluctuations are observed for all the measurements in the energy range below 2 MeV: variation in the peaks differ from each other that requires further investigation. The spectrum averaged γ-ray energy $<\varepsilon_\gamma>$, calculated by experimenters within the region 0.1 – 10 MeV, is equal 0.913 ± 0.032 MeV [45].

The γ-ray spectra of S. De and co-workers published in 2018 [45] were not used in the present GMA analysis since this experiment seems to be a preliminary investigation of this team. Moreover, the spectral data are not compiled in EXFOR. We multiplied the obtained average gamma ray energy $<\varepsilon_\gamma> = 0.913 \pm 0.032$ *MeV by multiplicity* $M\gamma = 8.75$ *γ/f which results in the total prompt energy* $<E\gamma> = 7.989 \pm 0.280$ *MeV/f (see Table4.2).*

(19) In 2021, D. Pandit et al. reported the measurements of the prompt γ-ray spectrum from ^{252}Cf(s.f.) within the range 1 to 50 MeV at the underground laboratory at Jaduguda, India [46].

The experiment was carried out with the aim to measure the γ-rays above 25 MeV which is a challenging task due to both low probability of emission and a large cosmic-ray background. The experiment was performed at the underground facility at a 555 meters depth level of the mine that provided a natural reduction of the cosmic muon flux by a factor of more than 10^4.

The ^{252}Cf source had the intensity 12 μCi [46]. The first array of four BaF_2 crystals, each having a 3.5 ×3.5 cm^2 cross-sectional area and 5 cm length, was mounted as close as possible to the ^{252}Cf source to detect the gammas from fission events as well as to get the start trigger for time of flight (TOF). The second array of twenty-five BaF_2 detectors, each having 3.5 × 3.5 cm^2 cross-section and 35 cm long, was employed to measure the high-energy γ-ray spectrum at a distance of 25 cm from the source. These two detector systems allowed to detect the prompt γ-rays from the ^{252}Cf source by the γ-γ coincidence within the time window 70 ns.

The TOF measurement provided also the neutron-gamma discrimination of the events in the high energy spectrometer [46]. The pile-up events were rejected by pulse shape discrimination. The background events from cosmic muons were also recorded for the same duration by replacing the ^{252}Cf source with a ^{22}Na source having similar activity. The calibration of the detectors was performed using ^{22}Na and Am-Be radioactive sources. The absolute efficiency of the detector array was calculated from GEANT4 simulation [46]. The high-energy γ-ray spectrum was generated in offline by summing the energy deposition in all the 25 detectors where every detector was required to satisfy the cuts employed via the two-dimensional pulse shape discrimination and the prompt time gate.

D. Pandit et al. [46] compared the measured prompt γ-ray spectrum accompanying spontaneous fission of ^{252}Cf with the previous measurements of J. Kasagi [25], S. Luke [30], H. Van der Ploeg [31], D. Pandit [32], N. Eremin [33] and R. Vind [42]. They found that all these PFGS are in good agreement in the medium energy region 6 - 20 MeV. However, the measured γ-ray spectrum above 25 MeV, as stated by the authors [46], was about two orders of magnitude lower than J. Kasagi [25], H. Van der Ploeg [31], D. Pandit [32] (the previous measurement of the same team but at surface level), and more than an order of magnitude lower compared to N. Eremin [33], R. Vind [42] or S. Luke [30]. The authors attributed the difference between their own measurement and previous ones to the incomplete rejection of the cosmic ray background which leads to an overestimation of the γ-ray emission probability.

The prompt γ-ray spectrum is provided in EXFOR sub-entry 33167002 in numerical form by the authors of [46]. As can be seen in Fig. 4.2, the first points up to 4 MeV are systematically lower than most of the others. No information is reported about the absolute normalisation, unfolding and correction applied. The energy dependant uncertainty is given in an EXFIOR entry but without any comments and analysis of its sources. The given values of uncertainty are extremely small 0.1 – 3.5% for the γ-ray energies below 16 MeV. Considering this, we used PFGS of D. Pandit et al. [46] as a shape data and at energies $E\gamma \geq 5$ MeV. Uncertainty ± 10% was added to those provided in sub-entry 33167002 and they were interpreted as statistical ones.

TABLE 4.1. The known measurements of the prompt photon (γ- or X-rays) energy spectra from ^{252}Cf(s,f.) and their main parameters. The time window for the fast coincidence with fission event (f) registered either by the fission fragments (FF), γ-rays (γ) or neutrons (n) is given in parentheses. The experimental data compiled in EXFOR by digitizing of the plots in the referenced publications are denoted by superscript [d], other data were received there from experimenters in numerical format.

First Author	Year Lab	γ energy range, MeV	^{252}Cf strength, f/s	γ-ray detector	Fission fragments or γ flash detector	Measurement technique (time coincidence window)	Corrections, Comments	Reference EXFOR Entry
Prompt γ-ray fission energy spectra (PGFS) measurements (all Fission Fragments)								
A. Smith	1956 ANL	0.13 – 6.6	8.3E+01	single-and double-NaI(Tl)	gas scintillator	γ-FF coincidence	no information	[18] 14320.002 [d]
H. Bowman	1958 LLNL	0.09 – 2.75	3.3E+04	NaI(Tl)	double Frisch-grid ioniz. chamber	γ-FF coincidence	little information	[20] 14387.007 [d]
N. Ajitanand	1969 BARC	0.10 – 1.79	5.0E+03	Ø5×5 cm NaI(Tl)	Si surface barrier detector	γ-FF coincidence (≈ 1 μs)	pulse-height spectrum	[21] 33083.002 [d]
V. Verbinski	1973 IRT, Diego	0.14 – 10.0	n/a	Ø5.8×15.2 cm NaI with anti-Compton shield	Si surface barrier detector	γ-FF coincidence (≤ 10 ns), NaI anti-Compton shield	all necessary applied	[22] 14195.004 [d]
F. Dietrich	1974 LLNL	7.0 – 19.0	3.7E+04	Ø25.4×25.4 cm NaI with anti-Compton shield	ionization chamber	γ-FF coincidence, NE102 plastic anticoincid. shield	all necessary applied	[23] 14642.002 - 4 [d]
J. Kasagi	1989 Tokyo	6.0 - 155	3.9E+04	7 crystals BaF_2, 37 cm^2 ×20 cm	Ø12.5 × 5 cm NE-213	γ-γ coincidence		[25] 23740.002 [d]
Y. Pokotilovskii	1990 Dubna	20 – 160	7.0E+06	Ø15×15 cm NaI(Tl)	no detection	anti-Compton shield, 25 cm long B-polyethelene	upper limit is reported	[26] 41577.002
E. Sokol	1991 Dubna	0.43 – 5.04	n/a	8 crystals Ø3.6×14 cm $Bi_4Ge_3O_{12}$	Si semiconductor	γ-FF coincidence (≤ 4 μs)	PH distribution was converted in energy	[27] 41580013 [d]
S. Luke	1991 Washington	5.3 – 20.1	1.9E+06	Ø25.4×38.1 cm NaI(Tl)	Si-SSD NE-213	γ-FF coincidence and γ-γ coincidence; anti-coincidence shield	NaI efficiency, pile-up	[29], [30] 14544.002 [d]
H. Ploeg	1992 Groningen	2.1 – 37.1	1.2E+04	Ø25×35 cm NaI two 2.6 l BaF_2	eight 3.5×3.5×6.0 cm^3 BaF_2 for γ; four PPAC for FF	γ-γ coincidence for TOF; γ-FF coincidence for angle correl., plastic anti-coinc.	random γ-γ coincidence, background	[31] 23739.002 [d]
D. Pandit	2010 BARC	4.0 – 79.0	1.1E+05	48 crystals BaF_2	4 crystals BaF_2	γ-γ coinc.; TOF γ-n discrim.; PSD pile-up rejection	insufficient information	[32] 33041.002

First Author	Year Lab	γ energy range, MeV	^{252}Cf strength, f/s	γ-ray detector	Fission fragments or γ flash detector	Measurement technique (time coincidence window)	Corrections, Comments	Reference EXFOR Entry
N. Eremin	2011 Moscow	4.4 – 60.0	1.6E+06	two Ø7.6×7.6 cm BGO	Ø6 × 2 cm polystyrene for neutron detection	γ-n coincidence; TOF selection (20 ns)	random γ-γ coincidence, background	[33] - [35] 41551.003 [d]
E. Kwan	2012 LANL	0.20 – 5.32	7.4E+04	six Ø12.5×5 cm organic scin.	parallel plate avalanche counter	γ-FF coincidence	all necessary	[36] 14413.005
A. Chyzh	2012 2014,LANL	0.2 – 9.4 0.10 – 10.5	5.6E+03 5.6E+03	160 crystals of BaF_2 cover. 4π	parallel plate avalanche counter	γ-FF coincidence (≈ 30 ns)	all necessary	[37] 14315.002 [39] 14361.011
A. Oberstedt	2015 IRMM	0.10 – 9.0	8.6E+03	$LaBr_3$:Ce and three $LaCl_3$:Ce	CVD diamond	γ-FF coincidence (≈ 6 ns)	all necessary	[40], [41] 23197.010-013
R. Vind	2016 BARC	2.0 – 56.0	1.9E+05	Ø5.0×7.6 cm BiGeO	multi-wire proportional counter	γ-FF coincidence (≈ 20 ns)	muon contamination	[42] 33165 [d]
L. Qi	2018 Orsay	0.09 – 8.01 0.10 – 8.04	n/a	seven $LaBr_3$Ce nine Phoswich	fission ionization chamber	γ-FF coincidence (≈ 5 ns)	all necessary	[43] 23417.002 23417.003
S. De	2020 BARC	0.10 – 5.14	n/a	two Ø3.81× 3.81cm $CeBr_3$	fission ionization chamber	γ-FF coincidence	background, unfolding	[44] 33135.003
D. Pandit	2021 BARC	4.0 – 48.0	4.4E+05	25 3.5×3.5cm^2 ×35cm BaF_2	four 3.5×3.5cm^2 ×5cm BaF_2	γ-γ coincidence (70 ns); pile-up rejection by PSD	555 m underground	[46] 33167.002
Prompt γ-ray fission energy spectra (PGFS) measurements (for selected mass range of Fission-Fragments)								
P. Glässel	1989 Heidelberg	0.15 – 9.0	6.0E+02	162 NaI 4π crystal ball	F1: solid state det. F2: avalanche det.	γ-FF coincidence (8 ns)	γ-spectra were not unfolded	[50] not in EXFOR
A. Hotzel	1996 Heidelberg	1.0 – 10.0	4.0E+02	162 crystals NaI(Tl)	double Frisch-grid ionization chamber	γ-FF plastic anti-coinc. γ-FF coincidence	all necessary	[51] 22757.002 [d]
C. Badimon	1997 CENBG	0.0 – 13.0	n/a	EUROGAM II multidetector	photovoltaic cells SAPHIR	Compton suppression, TOF to suppress neutrons	preliminary data	[54] prelim results
D. Biswas	1999 INFN	3.0 – 8.0	1.0E+04	80 BGO 40 HPGe	40 HPGe	TOF to suppress neutrons		[55] 23598.004 (but no γ-ray)
Prompt X- and γ-rays fission energy spectrum or yield measurements (all Fission-Fragments)								
L. Glendenin	1965 ANL	0.003 – 0.060	3.0E+03	Ø2.5×0.3 cm NaI(Tl)	Si surface barrier semiconductor	X-FF coincidences (< 2 ns)	probably all	[57], [58]
R. Watson	1967 LRL, CA	0.010 – 0.045	4.1E+03	0.6 mm^2 × 3 mm Si(Li)	0.5 mm thick P-diffused Si semicon.	X-FF-e coincidences (0 - 93 ns)	probably all	[59]
R. Watson	1970 LRL, CA	0.010 – 0.045	6.3E+04	10 × 10 × 3 mm Si(Li)	FF: Si semicond. e: magnet + Si(Li)	X-FF-e coincidences (≈ 1.7 ns)	probably all	[60], [61]

First Author	Year Lab	γ energy range, MeV	^{252}Cf strength, f/s	γ-ray detector	Fission fragments or γ flash detector	Measurement technique (time coincidence window)	Corrections, Comments	Reference EXFOR Entry
E. Cheifetz	1975 LRL, CA	0.07 – 1.28	8.3E+03	1-35 cm^3 Ge(Li) 2 cm^3 Si(Li)	two Si plated semiconductors	X-γ-FF, γ-γ coincidences (< 2 ns)	probably all	[62], [63]
Total γ-ray fission energy spectra (TGFS) measurements without γ-f time coincidence (encapsulated Cf sources)								
L. Schänzler	1976 Münster	0.5 – 6.0	2.7E+08	Ø4.65×4.85 cm NE-213 scintil.	no	Unfolding with response matrix	not published	shown in [66]
D. Ingersoll	1977 Urbana, IL	1.0 – 5.8	n/a	Ø4.65×4.85 cm NE-213 scintil.	no	Unfolding with response matrix	few information	[67] not in EXFOR
S. Jiang	1977 KFK	0.3 – 3.0	1.5E+07	Si(Li) Compton spectrometer	no	Unfolding with response matrix		[68], [69] SINBAD
L. Trykov	1977 IPPE	0.5 – 2.0	2.7E+08	Ø4.0×4.0 cm stilbene scintil.	no	Unfolding with response matrix		[70] - [72] ICSBEP
T. Czakoj	2023 Rez	0.20 – 14.3	3.4E+07	Ø4.5×4.5 cm stilbene scintil.	no	Unfolding with response matrix	all corrections	[74]

TABLE 4.2. The known measurements of the average prompt γ-ray multiplicity Mγ(p) and total γ-ray energy Eγ(p) emitted per fission of ^{252}Cf(s.f.). The energy ranges (from threshold to high-energy limit), where the γ-rays were measured, the average parameters and emission (coincidence) time are listed.

First Author, Lab (Year)	Threshold, keV	High-energy limit, keV	Coincidence time, ns	Mγ(p), γ/f	Eγ(p), MeV/f	Ref.	EXFOR
Prompt: γ-rays were registered in time coincidence with fission event (f)							
A. Smith, ANL (1956)	60			10.3 ± n/a		[18]	14320.004
H. Bowman, LNLL (1958)	40			10.0		[20]	14387.006
G. Val'skii, INP (1969)	100			7.50 ± 1.50 (20%)		[75]	
K. Skarsvag, IAK (1970)	100		0 - 120	11.6 ± 1.0 (8.6%)	6.99 ± 0.30 (4.2%)	[76]	23174004
F. Pleasonton, ORNL (1972)	85			8.32 ± 0.40 (4.8%)	7.06 ± 0.10 (4.4%)	[78]	14214.009
H. Nifenecker, Saclay (1972)	n/a				6.5	[79]	
G. Mehta, Saclay (1973)	n/a				7.0	[80]	23213.006
V. Verbinski, IRT (1973)	140			7.80 ± 0.30 (3.8%)	6.84 ± 0.10 (4.5%)	[22]	
E. Nardi, Soreq (1973)	25				6.7 ± 0.4 (6.0%)	[81]	
G. Brunson, LANL (1982)	85 140			8.32 7.80		[82]	
E. Sokol, JINR (1991)	320	5120		6.58 ± 0.46 (7%)	9.6 ± n/a (n/a)	[27]	41580.003
L. Krupa, JINR (2004)	100	6500		8.1 ± n/a	6.7 ± n/a (%)	[83]	23814.003
A. Chyzh, LLNL (2012) (2013) (2014)	150 150 150			8.14 ± 0.40 (4.9%) 8.16 ± 0.40 (4.9%) 8.75 ± 0.44 (5.0%)	0.96*8.14=7.81 8.52 ± 0.43 (5%)	[37] [38] [39]	
A. Oberstedt, IRMM (2015)	100			8.29 ± 0.13 (1.6%)	6.65 ± 0.10 (1.5%)	[41]	23197.009
T. Wang, Beijing (2016)	0[1)]			10.1 ± 1.0 (10%)		[87]	
L. Qi, IPN Orsay (2018)	90 116	8006 8039		8.30 ± 0.15 (1.8%) 8.40 ± 0.19 (2.3%)	6.60 ± 0.15 (2.3%) 6.70 ± 0.26 (3.9%)	[43]	23417.004
S. De, BARC (2018)	100	10000			7.99 ± 0.28 (3.5%)	[45]	
Total: all γ-rays without detection of fission event (f)							
L. Schänzler, Münster (1976)	600	6020	∞	5.460	1.500	[66]	
D. Ingersoll, Urbana (1977)	900	5850	∞	4.260	1.793	[67]	
L. Trykov, IPPE (1976) (1985)	350 407	 8900	∞	12.019 14.445	1.205 1.022	[70], [71] [72]	
S. Jiang, KFK (1977)	342	3000	∞	15.48 ± 1.50 (10%)	0.876	[68], [69]	
M. Kostal, CVŘ Řež (2013)	150	8000	∞	10.49 ± 1.00 (10%)	0.974	[74]	

Note: 1) T. Wang measured Mγ by γ-detector with 40 keV threshold as a function of FF total kinetic energy and linearly extrapolated to zero [87].

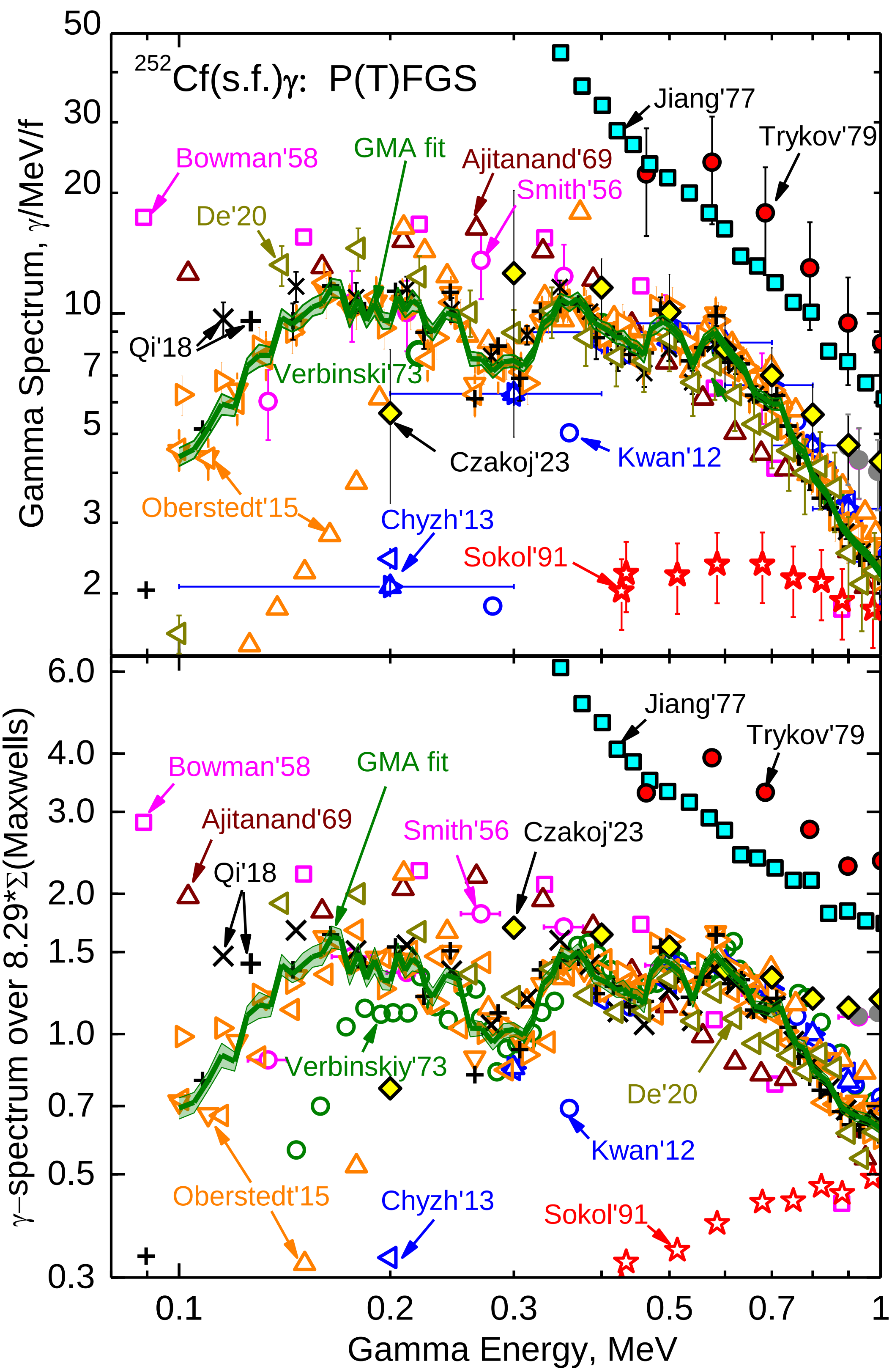

FIG. 4.1. γ-ray spectra from [252]Cf(s.f.) (top) and ratio over the sum of two Maxwellian distributions (bottom) in the energy interval 0.08 – 1 MeV. Experiment: prompt γ-ray emission (open symbols) and total (closed symbols). GMA evaluation of PFGS carried out in the present work: green curve with uncertainty as bar corridor.

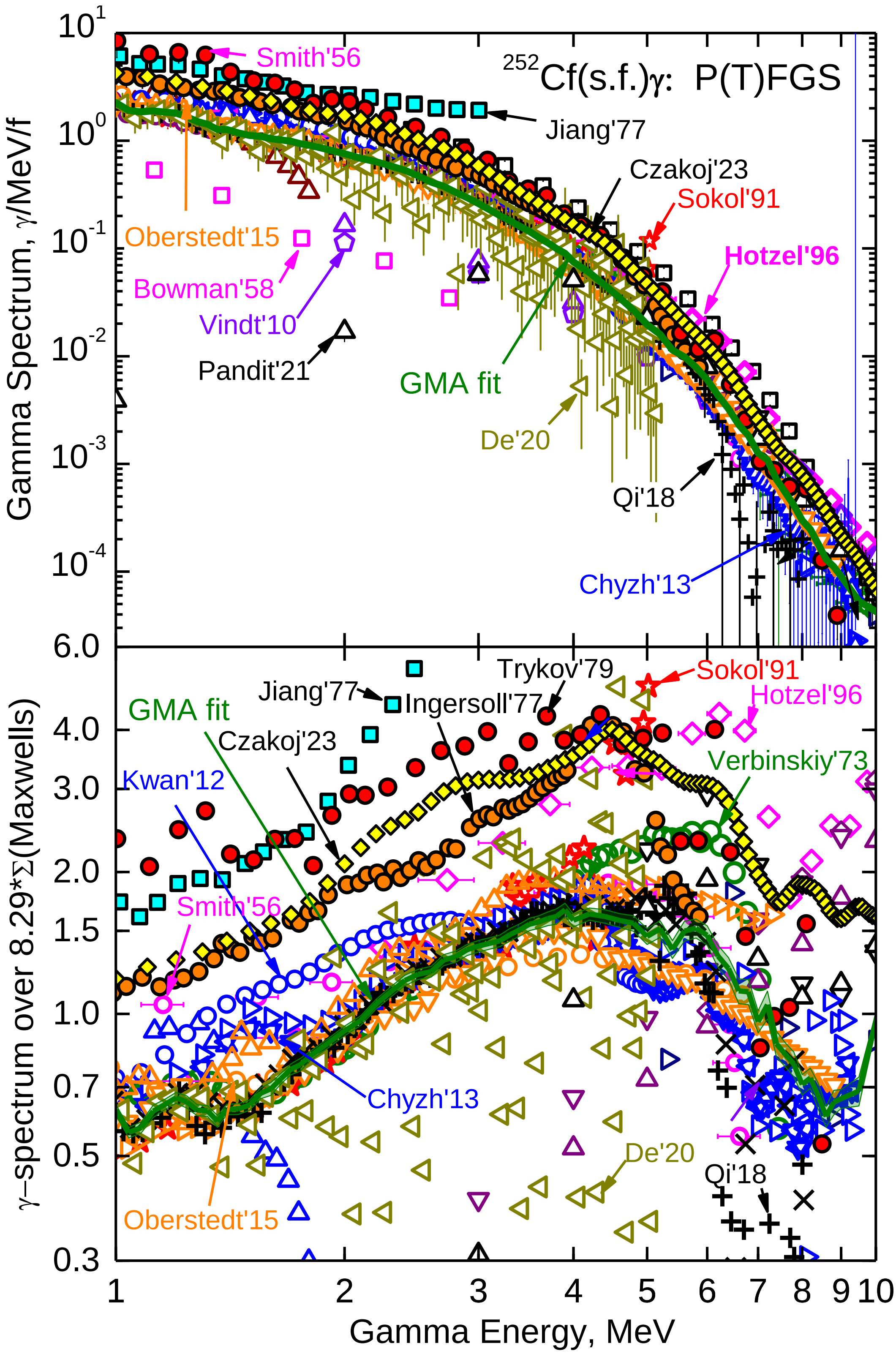

FIG. 4.2. γ-ray spectra from ^{252}Cf(s.f.) (top) and ratio over the sum of two Maxwellian distributions (bottom) in the energy interval 1 – 10 MeV. Experiment: prompt γ-ray emission spectra (open symbols) and total (closed symbols). GMA evaluation of PFGS carried out in the present work: green curve with uncertainty as bar corridor.

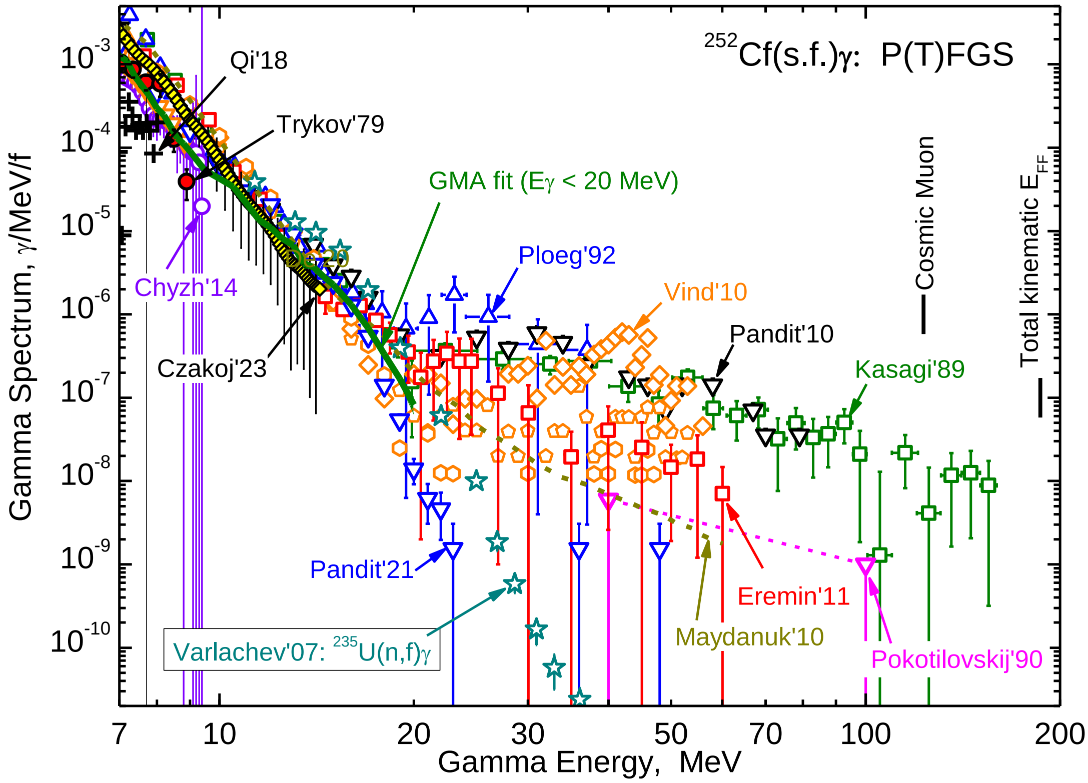

FIG. 4.3. Prompt γ-ray energy spectra from ^{252}Cf(s.f.) in the energy interval 7 – 200 MeV. Experiment: prompt γ-ray emission (open symbols) and total (closed symbols). GMA evaluation of PFGS carried out in the present work is a green curve with uncertainty as bar corridor. For comparison, PFGS from reaction ^{235}U(n_{th},f)γ [88] and calculated TFGS from ^{252}Cf(s.f.) [92] are co-plotted.

4.3. Measurements of the prompt fission γ-ray spectra with assignment to fission fragments

Several experiments could be found in the literature where the measured prompt γ-ray spectra were attributed to a certain fission fragment mass or range (split). The more advanced technique used in these works allowed to detect not only the fission fragments but also to measure their mass or kinetic distributions. Generally, such experimental data also allow to obtain the aggregate PFGS for ^{252}Cf(s.f.), if the γ-spectra are measured for practically all fragments. In this case the individual fission mass spectra should be weighted with independent mass yields, which is rather well known for the spontaneous fission of ^{252}Cf, and then summed.

Here we consider the results of four experiments. Several others (e.g., see [49]) could be found in the literature, but they are even less informative for the purpose of the present evaluation. Even among these four measurements we managed to derive the γ-ray spectrum, summed over the whole fragment masses, only for a single experiment. It was impossible to do this for the other ones due to insufficiency of the measured or reported information.

(1) In 1989, P. Glässel et al. reported the γ -ray spectra and multiplicities from ^{252}Cf spontaneous fission [50]. The experiment was performed with the Heidelberg-Darmstadt Crystal Bal1, a 4π detector with up to 162 NaI(Tl)-crystals and high efficiency for gammas and neutrons [50]. The ^{252}Cf fission fragments were measured in coincidence between a solid-state detector and an array of 7 low-pressure position-sensitive parallel plate avalanche detectors with ~ 1.4 π solid angle. The ^{252}Cf source with an activity of 600 fissions/s on the 0.22 μg/cm^2 polypropylene foil was mounted in the centre of the Crystal Ball at 1 mm distance from the solid-state detector. For fragment 1, the kinetic energy and time of arrival were measured in the solid state detector. For fragment 2, the direction and time-of-flight (with respect to fragment 1) over typically 20 cm were measured within 18 mrad. After all corrections, the overall resolution 400 ps for time-of-flight and ~ 1 MeV for kinetic energy, resulting in a (pre-neutron) mass resolution of 2.7 to 3.3 mass units, were achieved.

With a time resolution of the Crystal Ball of ~ 3 ns and 35 cm flight path, the prompt gammas were selected in the time window -4 to 4 ns. The remaining neutron contamination was less than 1%. The lower cut-off on the γ-energy was 150 keV [50].

The experimental results of P. Glässel et al. [50] include:

- the γ-ray energy spectra in the energy range 0.15 to 9 MeV for five selected mass splits (the light fragments range from 90 to 128). Because of the low statistics, the experimenters did not unfold the measured spectra for the gamma detector response since this was not important for a comparison of different mass splits;
- the relative γ-ray multiplicity Mγ, corrected for efficiency and pile-up, versus the individual fragment mass.

Neither uncorrected energy spectral nor relative multiplicity data can be used in the present evaluation of the reference ^{252}Cf(s.f.) spectrum.

(2) In 1996, A. Hotzel, P. Thirolf et al. measured the γ-ray spectrum of ^{252}Cf(s.f.) with the Darmstadt-Heidelberg crystal ball spectrometer and a double ionization chamber mounted inside to detect the fission fragments [51]. The crystal ball was made of 162 NaI-scintillator crystals and formed a sphere with an inner diameter of 50 cm. It served as a 4π γ-ray detector with high efficiency and angular resolution.

For the detection of the fission fragments, a double ionization chamber (DIC) was built into the crystal ball. The outer wall of the DIC was made of 1 mm aluminium and had an almost spherical shape with a diameter of 30 cm. The ^{252}Cf source had a diameter of 5 mm and a fission activity of 400/s. It was mounted on a thin backing (30 µg/cm^2 Al_2O_3, with 10 µg/cm^2 Au evaporated onto both sides) in the hole at the centre of the cathode. The cathode and symmetrically located on both sides two Frisch grids and two anodes had diameter 15 cm. The DIC allowed to measure the number of fission events within an active solid angle of 60% of 4π, the polar angle of fission fragments (FF) and their total kinetic energy (TKE). The authors corrected the FF mass values *A* referring to the fragments before the neutron evaporation. The fragment mass resolution (FWHM) was determined to be 5.0.

The NaI-detectors were calibrated with various calibrated γ-sources and with the aid of the two inelastic scattering reactions. The measured γ-spectra were corrected for the effect of gamma–neutron summing from the same fission event, which was quite strong for γ-energies above 4 MeV and reached 70% of the raw γ-spectrum at 7 MeV. The spectra were also corrected for γ–γ summing, the influence of which was, however, small. The measured γ-spectra were deconvoluted with the response matrix of the crystal ball. This matrix was created by a GEANT simulation of the bare crystal ball und renormalized according to the full energy peak efficiencies of the actual experiment, which were taken from the calibration measurements.

The numerical experimental data compiled in EXFOR were obtained by digitizing Fig. 11 in [51]. The author of the experiment [52] stressed that these data were obtained by summing over all emission angles and all total kinetic energies as well as over the two equivalent mass splits; they were unfolded from the crystal ball response matrix and then normalized to the number of fission fragments observed for the given mass splits. The mass values given to the right of Fig. 11 correspond to the lighter fragment mass A_L.

To obtain the total prompt energy spectrum of γ-rays from spontaneous fission of ^{252}Cf, we summed nine gamma spectra presented in Fig. 11 of [51] (and in EXFOR) weighting them with the independent fission yields taken from ENDF/B-VIII.0 [7]. Since the mass-symmetrized γ-ray spectra are given for the light masses A_L within masses split (A_i - A_{i+4}), the fission mass yields for every split was calculated as a half of sum of yields for the light and corresponding heavy fragment masses. Finally, the resultant spectrum was divided by 0.5 MeV since the data displayed in Fig. 11 are the gamma yields for energy interval 0.5 MeV [51].

The resultant mass integrated prompt γ-ray spectrum for ^{252}Cf(s.f.) is shown in Fig. 4.1 The ratio of experimental data over the sum of two Maxwellian distribution definitely shows that the data of

A. Hotzel et al. [51] start to systematically overestimate the bulk of known measurements at $E_\gamma \approx 3$ - 4 MeV reaching the factor ≈ 2 at $E_\gamma \geq 6$ MeV. The authors of [51] practically only discussed the statistical experimental uncertainties which are also depicted as the point bars in Fig. 11. Other uncertainties (e.g. for the response function calculated by GEANT4) were not quantified at that time [52]. Regarding this we assigned ± 10% systematic uncertainty for this data set.

One year later, P. Singer and co-workers published new measurements of the high-energy γ-rays emission from the binary and α-particle accompanied spontaneous fission of ^{252}Cf [53]. An experiment was again performed at the Darmstadt-Heidelberg Crystal Ball spectrometer. The spectra of prompt γ-rays in the energy range ≈ 0.2 to ≈ 10 MeV are presented as a plot in paper [53] for the heavy fragment mass splits with $126 \leq A_h \leq 136$ and $144 \leq A_h \leq 154$.

The mass splits which do not cover the whole fragment masses, the absence of information about unfolding, graphical representation, etc. prevented us to use the measured data of P. Singer et al. [53] for the present evaluation work.

(3) In 1996, C. Badimon et al. presented the preliminary measurements of the spectrum of γ-rays emitted in spontaneous fission of ^{252}Cf [54]. The γ-rays were recorded by the EUROGAM II multidetector, using the photovoltaic cells to detect fission fragments. The aim of the experiment was to investigate the γ-yield enhancement which appears between 3 and 8 MeV for the mass fragment ratio near 132/120.

The authors presented a plot with preliminary results for the γ-ray energy range 0 - 13 MeV for five heavy fragment mass splits between 126 and 156. Due to the preliminary character and too few information in the progress paper [54], these data cannot be used in the present analysis.

(4) In 1999, D.C. Biswas et al. reported the experiment performed at the gamma spectrometer in the Laboratori Nazionali di Legnaro [55]. The energy γ-ray emission accompanying the spontaneous fission of ^{252}Cf was measured in coincidence with individual fission fragments selected by discrete γ-ray transitions.

A sealed ^{252}Cf source (~ 10^4 f/s) was placed in the centre of the inner ball of 80 bismuth germanate scintillators (BGO) of a 6 cm thickness. An outer array consisted of 40 large volume Compton suppressed germanium detectors (HPGe), positioned in 7 rings at different angles. In the present experiment, the energy and time signals of the individual BGO scintillators were recorded in order to detect γ-rays up to ~ 20 MeV.

In the off-line analysis prompt HPGe γ-ray events were selected by requiring a gate of ≈ 40 ns in the TOF spectra started by the inner ball [55]. The selection of a single fission fragment was performed by gating on low energy γ-ray transitions in the HPGe spectra. These transitions are discrete γ-rays between low lying states in fission fragments.

Experimenters [55], as an example, presented the plot with the γ-ray energy spectra up to 11 MeV in coincidence with light and heavy fission fragment isotopes. They also noted with importance that the spectra and the relative yield data reported in their work were not corrected for the detector efficiency.

Based on the information above, the ^{252}Cf(s.f.) fragment mass dependent γ-ray energy spectra of D. Biswas et al. [55] cannot be used for evaluation of the reference PFGS.

4.4. Measurements of the prompt fission X-ray and γ-rays below ≈ 100 keV

The previous sub-Sections dealt with the numerous experiments which reported the measured energy distributions of the prompt γ-rays with energies more than ≈ 100 keV. This low energy limit is explained by the typical threshold of the detectors and technique used. Moreover, below 100 keV the X rays appear additionally to the low energy γ-rays. The spectroscopy of the mixed radiations of such energies requires specific technical equipment and methods.

The X-ray emission is caused by the electromagnetic radiation transitions in the electron shells of the fission fragments. An internal conversion of the γ-rays is the prior mechanism for producing two K and eight L electron vacancies. Approximately 5% of the γ-ray transitions are internally converted into X-rays [56]. This was confirmed by calculations which used the known data for mass and charge distributions of fission fragments, prompt γ-ray yield, internal conversion coefficients, K X-ray energy and fluorescent yield [57]. X-ray energies are characteristic of atom charge Z, their yields depend considerably on Z and A of the particular fragment. Since the vacancies arise from internal electron conversion, the X-rays are delayed by time corresponding to the lifetime of the nucleus excited state which emits γ-ray (the restructuring of the electron shells is substantially faster) [57].

We found in the literature several publications dealing with the experimental study of the low energy radiation accompanying spontaneous fission of ^{252}Cf (see [56] - [65] and references there). Regrettably, most of the papers report the measured radiation spectrum only in the restricted energy range, in the form of apparatus spectrum or in arbitrary units. Often, the studies provide only the specific parameters of radiation (e.g., individual transitions for the restricted fission fragments). The only experiment which provided the aggregate continuous energy spectrum of low energy prompt photons is of L. Glendenin et al. [57], [58]. The other three considered here (R. Watson et al. in 1967 [59] and in 1970 [60], [61]; E. Cheifetz and J.B. Wilhelmy with co-workers [62], [63]) reported the yields of the prompt discrete radiation transitions. These measured data are surveyed in this section in detail in an attempt to get a full picture about the energy spectrum of the prompt X- and γ-rays below ≈ 100 keV. The key parameters of experimental set-ups and methods are summarised in Table 4.1.

(1) In 1965, L. Glendenin and H. Griffin measured the yields, energy and time distributions of the K shell X-rays in spontaneous fission of ^{252}Cf [57], [58]. The Cf source with an activity 3 10^5 f/s was deposited on mica backing of sufficient thickness to stop all fission fragments (FF). Fission events were selected by a surface-barrier semiconductor detector (SSD) which subtended the solid angle nearly 2π. The X-ray energy spectrum was measured by a 25 mm in diameter and 3 mm thick NaI(Tl) placed coaxially with the source and fission detector, or in “180° arrangement” [57]. The absolute detector efficiency was determined with a set of sample sources. A time coincidence circuit selected (X-ray - FF) events within the resolution time 300 ns.

The time distribution of K X-ray emission was studied by delayed coincidence and time-of-flight methods. Using the "180° arrangement” and a coincidence resolving time of 60 ns, the delayed coincidence data for K X-rays in the energy region 10 – 45 keV were obtained. Three components of delayed radiations with half-life ≈ 10 ns (85%), ≈ 30 ns (7%) and ≈ 100 ns (8%) [57] were observed.

The limited time of the coincidence circuit necessitated the experimenters to use the time-of-flight technique for shorter times. In these experiments the X-ray detector viewed a beam of fragments during various portions of their total flight (FF spends ~ 2 ns for 2 cm) from source to fission detector. Copper collimators were used to define the field of view with a time resolution as short as 0.1 ns. By this method it was found that (30 ± 10)% of the X-rays are emitted in the first 0.1 ns and another (30 ± 10)% in the interval 0.1 to 1 ns [57]. The time distribution of the K X-rays with energies 10 – 45 keV was also studied in the work [57] using a 2π gas-flow detector operated in slow coincidence (3.5 μs) with a surface-barrier detector placed in the base of the gas chamber. Beryllium, aluminium and gold absorption curves were constructed in order to distinguish electrons from low energy photons. These measurements indicated that, on average, (1.0 ± 0.2) electron (60 keV < E < 300 keV) was emitted per fission [57].

L. Glendenin and H. Griffin reported the energy spectrum of pure K X-rays in the region 5 to 60 keV. An effect of the prompt γ-rays was removed by measuring the background with a copper filter. The background dominated below 10 keV and above 50 keV. The authors [57] observed two maxima at 18 and 32 keV in the X-ray spectrum which correspond to the light (atomic number Z ≈ 43) and heavy (Z ≈ 55) mass groups. The spectrum was corrected for transmission and detection efficiency. The average yield of 0.55 ± 0.05 K X-rays per fission was reported.

The measured net K X-ray energy spectrum is presented in Fig. 1 of paper [57]. *We digitised these data and plotted them in Fig. 4.4 after the following correction. The area under the digitized spectrum was found to be 0.172 X/f. Since it is not clear whether the data plotted in Fig. 1 of [57] were absolutely normalized, we increased the digitized spectrum by a factor 0.55 / 0.172 = 3.20.*

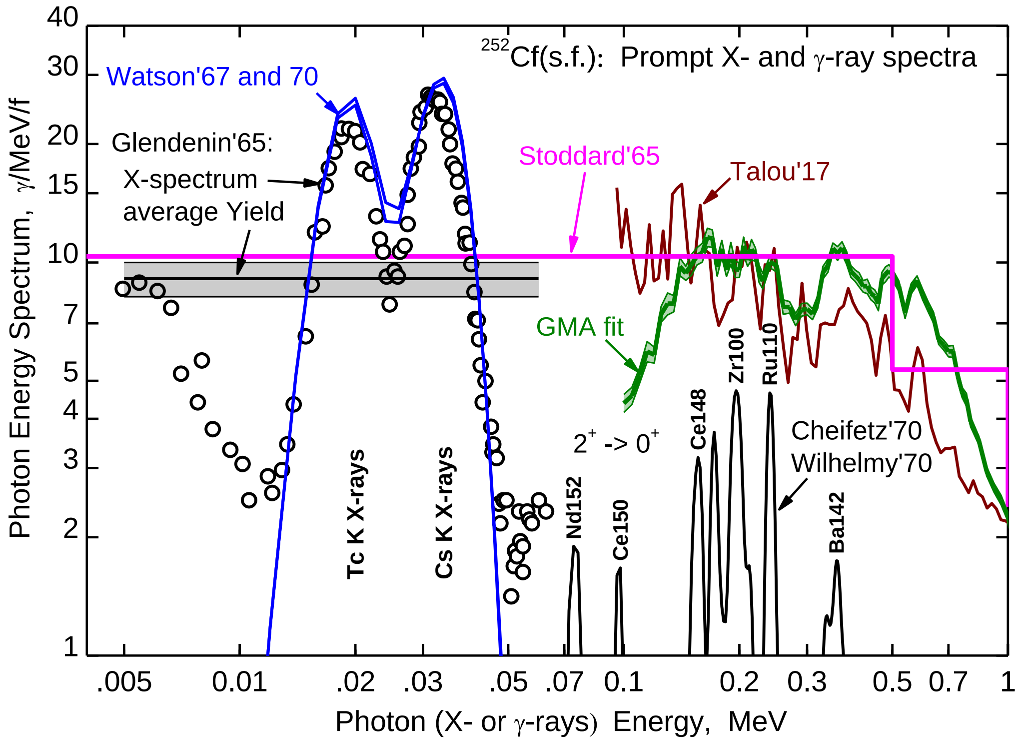

FIG. 4.4. Prompt X-ray and γ-ray spectra from ^{252}Cf(s.f.) in the energy interval 3 keV – 1 MeV. Measurements: L. Clendenin [57] (black open circles) - prompt K X-ray emission spectrum after renormalization (see text), black horizontal bar with grey uncertainty corridor – the average spectrum obtained from reported yield [57]: (0.55 ± 0.05)/(0.060 - 0.005) X/MeV/f; R. Watson et al. (blue curve) - prompt K X rays measured in 1967 [59] and 1970 [60], [61]; E. Cheifetz and J. Wilhelmy [62], [63] (black curve) - prompt K X-ray. The dominating K X-ray energies from light ($_{43}$Tc) and heavy ($_{55}$Cs) fission products and γ-rays from $2^+ \rightarrow 0^+$ transitions in several even-even fission isotopes (^{152}Nd - ^{142}Ba) are properly labelled.
Theory: brawn curve - prompt γ-ray spectrum calculated by P. Talou [90], [91]. Evaluation: pink histogram - prompt γ-ray spectrum of D. Stoddard [13], [14]. GMA evaluation of PFGS carried out in the present work – green curve with uncertainty as bar corridor.

(2) In 1967, R. Watson et al. measured the yields of prompt (pre β-decay) K X rays emitted by primary ^{252}Cf fission products [59].

The X-ray energies were measured in this experiment with a Si(Li) semiconductor spectrometer having dimensions 0.6 mm^2 × 3 mm and relative energy resolution ΔFWHM / FWHM = 0.82 keV / 26.25 keV ≈ 3%. A ^{252}Cf source (fission rate 2.43 10^5 fissions per minute) was mounted on a 90 μg/cm^2 Ni foil and was separated from the X-ray spectrometer by 0.020 inches of beryllium and 0.002 inches of aluminium. A 0.5 mm thick phosphorus-diffused silicon detector for counting fission fragments (FF) was located behind the aluminium stopping plate and close to the Cf source to ensure that all fragments were stopped within 5 10^{-11} ns after fission. Timing pulses from the X-ray and FF detector amplifiers were used to select coincidence within the time interval of 0 to 93 ns after fission in order to generate a gating signal. The accidental coincidence rate was measured to be less than 1% of the total coincidence rate. The X-ray spectrometer efficiency was calibrated by measuring the intensities of the 13.9, 17.8 and 20.8 keV X rays, 26.35 keV and 59.54-keV γ-ray from a calibrated ^{241}Am source and of the 32.2, 36.4-keV X rays from ^{137}Cs. The X-ray K_α and K_β groups contributing to the primary ^{252}Cf fission products were not fully resolved. The least-squares peak-fitting procedure was applied in order to determine accurate values of the X-ray intensities for each element.

The experimenters presented the yields of K X-rays per fission in a figure and listed them in a table as a function of atomic number [59]. They observed a maximum in heavy fission-product X-ray yield occurring at Caesium (Z = 55), and the maximum in the light-product at Technetium (Z = 43). Summing the yields of all the light-fission, heavy-fission and total-fission-product K X rays, the authors received 0.205 ± 0.005, 0.375 ± 0.008, and 0.58 ± 0.01 X-rays/fission, respectively. These values were in satisfactory agreement with corresponding data reported by L. Glendenin et al. [57], [58] and others, thus providing a check of accuracy.

We took the absolute intensities of the K X rays for 25 light and heavy fragments of ^{252}Cf(s.f.) from Table 1 and their energies from Figs 2 and 3 published in [59]. They were folded with Gaussian distribution. To get the X-ray energy distribution visually comparable with L. Glendenin et al. [57], we increased the relative energy resolution original value 3% to 30%. The resulting energy spectrum is depicted in Fig. 4.4.

(3) Later in 1970 R. Watson et al. reported the results of multiparameter measurements of complementary fission fragments, prompt K X rays, conversion electrons, and γ rays from spontaneous fission of ^{252}Cf [60], [61].

A ^{252}Cf source with a fission rate of $\approx 3.8\ 10^6$ fissions per minute was deposited onto a ≈70 μg/cm^2 nickel foil and was mounted coaxially between two phosphorus-diffused silicon fission fragment (FF) detectors [60], [61]. The fragment detectors were collimated to Ø15 mm and operated at - 50'C. One fission fragment detector (Fl) was located 1.0 cm to the left of the fission source and the other (F2) 2.0 cm to the right. A lithium drifted silicon X-ray detector of dimensions 1 cm × l cm × 3 mm was mounted in a separate evacuated cryostat which was isolated from the main vacuum chamber by a 0.025 cm beryllium window and was located at 2.0 cm below the FF detector - Cf source axis. The X-ray detector and internally mounted field-effect transistor were operated at liquid nitrogen temperature and gave an energy resolution FWHM = 0.75 keV at 14 keV.

The electron spectrometer (a lithium-drifted silicon detector of dimensions 1 × 2 × 3 cm) was located inside the fission chamber [60]. The electrons were guided around a 90º away from the Cf source by the magnetic steering device to a shielded detector. The electron detector was operated at liquid nitrogen temperature and had an energy resolution of 2.5 keV FWHM for the 661 keV electron peak from the ^{137}Cs decay. The arrangement enabled an observation only electrons which were emitted during ≈ 1 ns after fission with a time resolution FWHM = 1.7 ns.

From the analysis of the triple coincidence electron measurements (e-F-X), the numerous low-energy conversion electron and K X-ray transitions and half-lives were assigned to fission products by experimenters [60]. Thus, they approximated the decay curve of the total yields of electrons (summed over all energies) by two component decay with the half-lives 0.17 and 2.6 ns. This confirms the definition of the prompt radiation as one happening within the first 3 ns after fission, which was introduced in earlier works [57], [59].

The authors [60], [61] unfolded complex FF mass-energy sorted X-ray spectra and eventually determined the most probable charge and mass values for the emission of K X-rays. The final results, the K X-ray intensities per fission as a function of fragment mass interval and atomic number, are presented in Table I of [61]. The measured multiplicity of the K X rays is 0.575 X/f with total uncertainty ± 20% [61]. *We took from there the total (summed over isotopes of the same element) intensities for 10 light and 10 heavy elements. The X-ray energies were assigned in the same manner as the earlier Watson' data [59]. After folding with 30% relative energy resolution the resultant energy spectrum was plotted in Fig. 4.4, where it can be seen that it well agrees with the one derived from the previous experiment of Watson et al. in 1967 [59].*

(4) E. Cheifetz and J.B. Wilhelmy with co-workers [62], [63] reported the yields of the prompt discrete γ-rays and/or associated K X rays belonging to the ground state bands of light and heavy fission products produced by ^{252}Cf(s.f.).

The authors performed three separate experiments using different photon detectors [62], [63]: (1) recording γ rays with a 1-cm^3 Ge(Li) detector (resolution 1 keV at 122 keV); (2) recording γ rays and/or X rays in coincidence using a 6 cm^3 Ge(Li) detector and a 2 cm^2 Si(Li) detector; and (3) recording γ-γ coincidences with a 35 cm^3 Ge(Li) coaxial detector and a 6 cm^3 Ge(Li) detector. In all these experiments a ^{252}Cf source of a nominally 10^5 fissions/min was electrodeposited onto the surface of the first fragment detector F1. The Doppler shifting and broadening problems were eliminated for transitions from the fragments stopped during the time ≈ 1 ps in that detector. The second solid-state detector F2 was separated by 8 mm from the first. The γ-lines associated with this detector were sharp and unshifted if they were emitted after the fragment arrived at F2 but were broadened and shifted when they were emitted by the fragment in flight. The lifetimes in the time region 0.1 - 2.0 ns were obtained from the ratio of the non-Doppler shifted γ-ray intensities observed when the fragments stopped in two separated detectors.

The experimenters [62], [63] calculated the fragment masses from the measured energies. Four γ-ray spectra associated with fragment masses in mass intervals 2 atom mass units (amu) wide were obtained by sorting the three-parameter data. Each of these spectra were then analysed to derive the values of energies and intensities of individual transitions. The X-ray, γ-ray coincidence data were used to obtain definite Z assignments for the observed transitions, whereas the γ-γ coincidence data – information on additional transitions associated with single isotopes.

The results of the investigation are summarized in tables of [62] (light fragments) and [63] (heavy fragments). The experimenters listed the energies and intensities (after correction for internal conversion) per ^{252}Cf(s.f.) for transition from the first 2^+ excited to 0^+ ground states, as well as assigned isotopes and level half-life (0.2 – 2.7 ns). All even-even isotopes with prompt yields more than 1% were identified. Regrettably the γ-ray intensities for the higher excited states in these bands were not reported.

We took the energies and absolute intensities of the $2^+ \rightarrow 0^+$ transitions in 12 light fragments and 16 heavy fragments. The resultant energy spectrum after folding with relative energy resolution 5% is depicted in Fig. 4.4.

It is seen that measurements of L. Glendenin [57] and of R. Watson (both in 1967 and 1970) [59], [60], [61] provide agreeing results for the spectrum of the X-rays within the energy range 10 to 50 keV. No other complete measurements of the energy distributions below ≈ 100 keV or discrete yields as well as the theoretical predictions seem to exist. L. Glendenin and R. Watson drew the following conclusions clarifying the physics of the X- and γ-rays emission from ^{252}Cf(s.f.) with energies below 60 keV: (i) contribution of the γ-rays is small due to complete conversion into electrons (the internal electron conversion coefficient is close to unity [57]); (ii) the main contributors are the fission fragments Tc and Cs and nearby elements; (iii) the time of the prompt radiation emission is within the first 2 – 3 ns after fission.

It is also interesting to notice that the X-ray yield measured in the energy interval 5 – 60 keV, (0.55 ± 0.05) X/f [57], (0.58 ± 0.01) X/f [59] and (0.58 ± 0.12) X/f [61], confirms the yield 0.575 X/f in the same energy range 5 - 60 calculated from evaluation of D. Stoddard [13], [14]. It is worth remembering that the latter has the single energy bin from 0 to 500 keV.

At higher photon energies, ≥ 60 keV, only E. Cheifetz and J. Wilhelmy [62], [63] measured the discrete X- and γ-ray yields. It is obvious that this experiment delivered only the fraction of all gammas, since only the γ-rays from depopulation of the lowest 2^+ states in the even-even fission fragments were reported. This is well confirmed in Fig. 4.4, where the energy spectrum derived from the discrete data of E. Cheifetz and J. Wilhelmy underestimates the GMA evaluated PFGS in the energy range 100 – 400 keV by a factor of 2 to 3. However, we can derive an informative message too: the several discrete γ-lines measured by E. Cheifetz and J. Wilhelmy, as can be seen in Fig 4.4, correlate with prominent peaks in the GMA evaluated prompt spectrum, that clarify their origin.

Summarising this Section, we conclude that experimental information about prompt radiation from ^{252}Cf(s.f.) is rather scarce and incomplete which makes the non-model evaluation of PFGS relying on the measured data below 100 keV impossible. The prompt means within first 2 – 3 ns: this decay time was either directly measured or was known as the half-lives of the identified discrete levels in the fission fragments. The existing experimental results for the X-ray spectrum below 50 keV, on average, seem to confirm the D. Stoddard evaluation. Thus, the latter could be used for extrapolation below 100 keV, at least for the purpose of the prompt multiplicity estimation in the whole energy range, see Section 5.

4.5. Measurements of the total fission γ-ray spectrum

So far five experiments have been carried out, where the Total Fission Gamma Spectrum (TFGS) from spontaneous fission of ^{252}Cf was measured. Since the detection of fission fragments is not foreseen in such experiments, the used Cf sources are usually stronger than in the γ-f coincidence measurements described in Section 4.2. The larger amount of californium was usually distributed in a substrate surrounded by one or two capsules. The set-ups include only the γ-rays spectrometers to measure the total emission of the γ-rays and their energy distribution. The absolute normalization of the reported gamma spectra is typically done per one neutron following the spontaneous fission of ^{252}Cf. The neutron intensity of the Cf sources was either obtained from the source producer or was measured in the respective lab. For the present evaluation we used the well-known value of the total neutron multiplicity Mn(t) = 3.7676 n/f [11], [7] for the renormalization of the γ-spectrum per one fission event (f).

The five known measurements of PFTS are individually described below, with special attention to the details which have an impact on the use in the GMA analysis.

(1) The group of scientists from Germany, L. Schänzler et al., was probably one of the first who measured the spectrum of γ-rays from the encapsulated Cf source. The experiment seems to have been an extension of the neutron spectra measurements with the same Cf source and NE-213 detector published by K. Clausen, ..., L. Schänzler et al. [66]. The spectrum within the γ-ray energy range from 0.5 to 6 MeV is plotted in Fig. 14 in D. Ingersoll et al. [67] and is cited there as a private communication with L. Schänzler in 1976.

Following the information presented in [66], the 50 µg of ^{252}Cf was contained in matrix substance within a stainless-steel capsule and iron tube of outer diameter 14 mm. The source had the strength (9.99 ± 0.14)·10^8 n/s and was mounted on a metal rod. The spectrometer system used by experimenters consisted of a 2 × 2 inches NE-213 detector and an electronic system for neutron and gamma-ray discrimination. The measured pulse-height distributions were unfolded by the FERDOR code. In [66] results are reported of measurements of the neutron spectra from the bare Cf source and transmitted through the liquid air. No information is given about the γ-ray spectra measurements.

We digitized the γ-ray spectrum from Fig. 14 of [67] and plotted it in Fig.4.1. It shows that the L. Schänzler data reasonably agree with the bulk of the other experiments. We used his γ-ray spectrum in our GMA fit at gamma energies Eγ > 1.5 MeV and as a shape. The statistical uncertainty was assigned to be ± 20%.

(2) In 1977, D.T. Ingersoll and B.W. Wehring measured the energy spectra of the total gamma rays from ^{252}Cf(s.f.), Na and Pu-Be sources [67].

No information about Cf source used is given in [67]. The NE-213 scintillation detector of 4.65 cm diameter by 4.80 cm length was employed for the spectroscopy of γ-rays. The pulse-shape discriminator reduced the counts from neutrons. The room background was measured with the Pb shadow bar placed between the Cf source and the detector.

The experimenters thoroughly investigated the γ-ray response of NE-213. Ninety-five γ-ray response matrixes were measured for NE-213 using a superconducting electron microtron combined with a bremsstrahlung monochromator. The results along with radio-isotope measurements were used to construct a gamma-ray response matrix between 0.8 and 11.5 MeV for unfolding the gamma-ray pulse height data. In order to test the response matrix, gamma-ray spectra from ^{24}Na, Pu-Be, and ^{252}Cf sources were measured and unfolded using the FORIST unfolding code [67].

The unfolded energy spectrum is shown as vertical error bars in figure 14 of [67]. The spectrum is compared to a similar measurement by L. Schänzler (obtained by the authors via private communication in 1976) and a measurement of the prompt γ-rays from ^{252}Cf made by V. Verbinski et al [22]. The authors stated [67] that their measurement is in good agreement with L. Schänzler's results, although generally 7 - 10% higher. They also noted that the small oscillation above 4 MeV probably is an artefact from the unfolding procedure and, even without the oscillation, their measurement would fall below L. Schänzler's results above 5 MeV.

Fig. 4.2. compares the total γ-ray spectrum from ^{252}Cf(s.f.) of D. Ingersoll [67] with all other known data. It shows that this data set indeed is slightly higher than the others and starts to drop above 5 MeV. Regarding these circumstances we used the D. Ingersoll' γ-ray spectrum from 1.5 to 4.9 MeV as a shape. The statistical uncertainty was assigned to be ± 20%.

(3) In 1977, S.H. Jiang et al. measured the total γ-rays energy spectrum from ^{252}Cf(s.f.) from 0.3 to 3.0 MeV [68], [69]. The Cf source consisted of ≈ 30 µg of ^{252}Cf distributed in a SiO_2/MgO matrix, which was encapsulated in inner and outer containers made of Zircaloy and Aluminium alloy. The inner container had a shape of hollow cylinder with inner sizes Ø6 × 4 mm and outer Ø10 × 10 mm. The outer container had a shape of hollow cylinder with sizes Ø11 × 11 mm and Ø15 × 15 mm. The Cf-source neutron strength was determined by three independent methods: comparison with a standard ^{252}Cf source; measuring by a calibrated ^{235}U fission chamber; and by proton-recoil neutron spectrometer. These calibrations resulted in the absolute neutron strength of the Cf-source: 5.51 10^{+7} n/sec with uncertainty ± 5%.

The experimenters [68] employed the Si(Li) Compton spectrometer to measure the γ-ray energy spectrum at distance 102.5 cm from the Cf source. The sensitive volume of the Si(Li) crystal had the form of a cylinder with an area 110 mm^2 and a thickness 5 mm. In this experiment neither a shield around the Si(Li) detector, nor a collimator were used to eliminate the room returned radiation toward the detector.

The Si(Li) detector response functions, i.e. the pulse-height distributions of Compton electrons produced by the monoenergetic γ-rays, were determined experimentally by using a set of monoenergetic gamma sources, which produced several discrete energy γ-rays from 0.511 to 2.754 MeV [68], [69]. For the construction of the complete matrix of the response functions, the Compton electron spectra were extrapolated to zero. The energy resolution of Si(Li) was found to decrease from 20 to 7% when γ-ray energy increases from 0.5 to 3.0 MeV.

The systematic errors of the derived γ-ray spectra were mainly due to errors in the response function. They included: accuracy of the ^{252}Cf-source neutron source strength (±5%); uncertainty of the absolute efficiency determination with a help of calibrated ^{137}Cs source (≈ 2%); energy dependence of the γ-rays absorption in detector walls (≈ 2%); extrapolation of the measured response function to zero energy (< 10%). The total uncertainty caused by the response functions was estimated to be ≈ ±10% which was derived from comparison of measurements with several calibrated gamma sources. Additional uncertainty resulted from the electron spectra unfolding, namely from neglecting the γ-rays contribution with energies above ≈ 3 MeV, which are produced by the neutron capture and inelastic scattering. Such influence of the high-energy γ-rays and corresponding systematic errors were found by varying the upper energy limit from 2.7 to 1.8 MeV. The authors [68], [69] concluded that the high-energy γ-rays will not disturb the measured γ-ray spectra below ≈ 1 MeV. Between 1 and 2 MeV, errors varying from ≤ 10% to ≤ 20% were expected. This latter uncertainty we linearly interpolated between 1 and 2 MeV.

An important warning is given by the authors of the KFK experiment: “quantitative interpretation of the measurements will be restricted to the energy region between 0.5 and 2 MeV" [68]. Fig. 4.2 confirms this tendency of too high yield of γ-rays above 2 MeV through the comparison with the γ-ray spectrum measured independently at IPPE up to ≈ 9 MeV [70].

The total γ-ray spectrum of S.H. Jiang is shown in Fig. 4.2 and could be compared with other known TFGS experimental data. As can be seen it covers a rather narrow energy range, moreover its shape visibly differs from other experimental data. Due to this reason the data of S. Jiang et al. were not considered in the actual GMA fit.

(4) In 1979, L.A. Trykov et al. measured the γ-ray spectrum with a stilbene scintillation spectrometer in the energy range 0.5 to 9.1 MeV [70] - [72]. This experiment was compiled in the ICSBEP handbook under entry “ALARM-CF-FE-SCHIELD-001” [72].

The highly intensive Cf source (444 µg, 1 10^9 ± 5% n/s) was manipulated with a fishing-rod with a magnet at the end. A radionuclide ^{252}Cf was distributed in the Al_2O_3 substrate of diameter 4 mm and height 3 mm. It was enclosed in the 12X18H10T stainless steel shell with external diameter 10 mm and height 8 mm. Additionally, the source was placed in a copper capsule 15 mm in diameter and 14 mm high with a wall thickness of 2 mm.

The γ-ray spectrum in the energy range 0.5 to 9.1 MeV was measured with scintillation spectrometer, consisting of the stilbene crystal Ø4.0 × 4.0 cm and photomultiplier [70] - [72]. An electronic scheme selected pulses induced by the Compton effect. Calibration of the energy scale and absolute efficiency of the gamma spectrometer was performed using a set of calibrated sources of photons whose intensities were known to ±2%. The detector was located at distance 70 cm from the Cf source. The shadow cone allowed to measure the background. The relative energy resolution was found to follow the law *R(E) = 15% / sqrt(Eγ [MeV])*.

The experimenters reported that statistical uncertainty is dominant. The systematic uncertainty of efficiency normalization was estimated to be 5% [70] - [72]. The total uncertainties are given in the ICSBEP entry [72] and range from 20 to 40%.

We used the γ-ray spectrum measured by L.A. Trykov et al. (numerical data are compiled in the ICSBEP handbook under entry "ALARM-CF-FE-SCHIELD-001" [72]) in the present GMA fit as a shape. Only the experiment data above 1.8 MeV were considered, since at these energies TFGS should be identical to the PFGS. We also corrected the measured energy spectrum from the encapsulated (or bare) Cf source for the multiple scattering and attenuation of the ^{252}Cf(s.f.) γ-rays and neutrons in the capsule and californium containing matrix [73]. These calculations were carried out by the Monte-Carlo technique employing the capsule model reported in [72] and the neutron and photon cross section from the ENDF/B-VIII.0 library. The gamma and neutron emissions from ^{252}Cf(s.f.) were sampled simultaneously, using the information about multiplicities and energy spectra available at that time [73]. The resulting correction function, which we used to multiply the measured bare Cf-source gamma spectrum to get the capsule-free spectrum, is depicted in Fig. 4.5. This correction results in a ≈10% increase in most gamma energies, thus compensating attenuation of γ-rays in Fe and Al. Except in the vicinity of 7 – 9 MeV: there the decrease by ≈40% obviously accounts for the prominent γ-lines from the neutron radiation capture in the Fe and Al of the capsule.

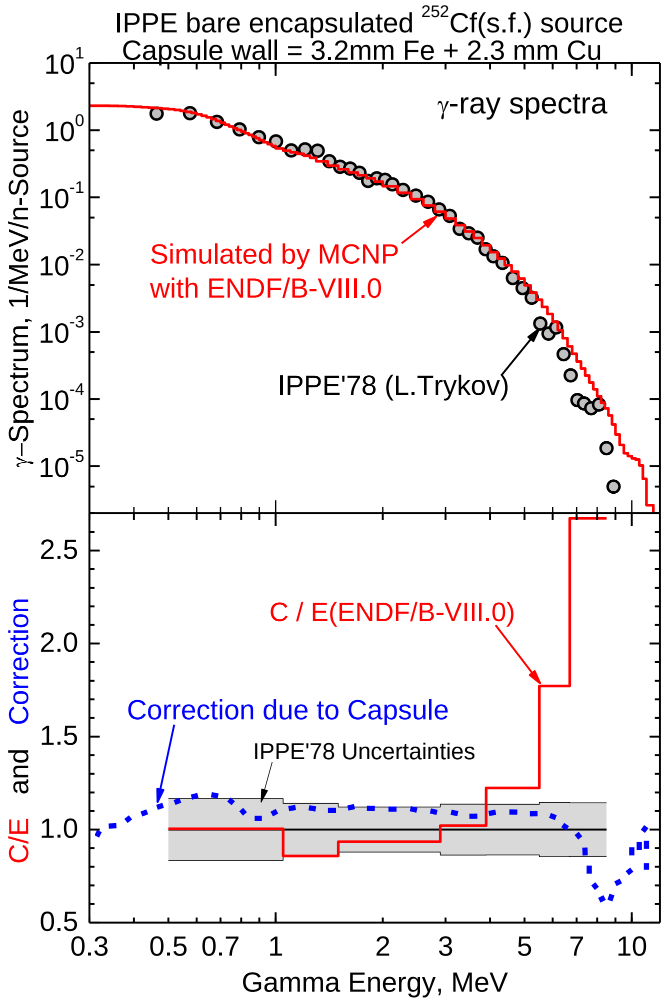

FIG. 4.5. Top. The total γ-ray energy spectrum from the IPPE encapsulated ^{252}Cf(s.f.) source: points - the IPPE experimental data of L. Trykov et al. [70] - [72]; red curve - MCNP simulation with the ENDF/VIII.0 neutron and photon cross section data.
Bottom. Ratio C/E (red histogram) and correction function (blue dashed curve) for the multiple scattering and attenuation in the IPPE encapsulated Cf-source. The grey corridor displays the total experimental uncertainties for the IPPE gamma spectrum.

(5) In 2023, T. Czakoj, M. Kostal et al. measured the total fission γ-rays energy spectrum in the energy range from 0.12 to 14 MeV [74]. They used a Cf source with intensity ≈ 1.2 10^8 n/s. The isotope ^{252}Cf in the form of compound Cf_2O_3 was dispersed in the Pd matrix of 3.9 mm diameter and 6 mm length, which was incapsulated in the 304L stainless-steel hollow cylinder having outer sizes Ø9.5 × 10 mm. The Cf capsule was transported in an aluminium tube to the measuring point inside the aluminium tube ending by the flexo-rabbit pneumatic system.

The energy spectrum of γ-rays was measured with the scintillator spectrometer consisting of a stilbene crystal of Ø4.5×4.5 cm and photomultiplier [74]. The gammas were well separated from the neutrons using the pulse shape discrimination technique. The gamma energy spectrum was obtained from the measured pulse distribution by deconvolution using the Maximum Likelihood Estimation, see details in [74]. The stilbene response matrices were calculated by the Monte-Carlo code and were additionally validated against the measured gamma spectra from the ^{24}Na and Am-Be sources.

The γ-ray detector was located at distance 100 cm from the Cf source. The 50 cm long lead shadow bar (cylinder of diameter 6 cm) was placed in between to measure the room scattered background. The measurement was carried out in a laboratory hall with dimensions 7.2 × 6.5 × 7.2 m at elevation 2 m above the ground. The detailed simulation with the MCNP6.2 code was performed by the authors of the experiment [74] to investigate and calculate corrections for the scattering and attenuation of gammas and neutrons in the materials surrounding ^{252}Cf in the laboratory hall. In the direction from the Cf substrate towards the γ-detector the thickness was about 2.8 mm of steel and 3 mm of aluminium. This resulted in the correction of the raw spectrum around 0.9 above 2 MeV and 1.5 at 0.2 MeV.

T. Czakoj, M. Kostal et al. [74] provided the absolute ^{252}Cf(s.f.) total gamma ray spectrum in the energy range 0.2 to 14.3 MeV. The following uncertainties were reported: response matrices of the stilbene detector and deconvolution of spectrum ≈ 5%. As can be seen in Figs 4.2 and 4.3 their data reasonably agree with other TFGS measurements and with PFGS but excluding the absolute scale. *For this reason, T. Czakoj et al. data were used in our GMA fit as a shape, the energy range was restricted at low energies by 1.8 MeV.*

4.6. Average Multiplicities and Energy of the prompt and total fission γ-rays

In several cases the experiments, where the prompt or total γ-ray energy spectra *S(E)* were measured, additionally provided the average γ-ray multiplicity *Mγ* or average γ-ray energies E_γ or ε_γ (see overview in Section 4.2 and data in Table 4.2). These quantities were usually obtained from the integration of the measured γ-ray energy distribution *S*(*E*):

γ-multiplicity per fission $$M_\gamma = \int_{E_{thr}}^{E_{max}} S\left(E_\gamma\right) dE_\gamma \quad (1)$$

average energy released in fission $$E_\gamma = \int_{E_{thr}}^{E_{max}} S\left(E_\gamma\right) E_\gamma\, dE_\gamma \;/\; \int_{E_{thr}}^{E_{max}} S\left(E_\gamma\right) dE_\gamma \quad (2)$$

spectrum average energy $$\varepsilon_\gamma = \int_{E_{thr}}^{E_{max}} S\left(E_\gamma\right) E_\gamma\, dE_\gamma \,/\, \int_{E_{thr}}^{E_{max}} S\left(E_\gamma\right) dE_\gamma = E_\gamma \,/ M_\gamma \quad (3)$$

If *S(E)* is an energy spectrum of the prompt γ-rays, then equations (1) – (3) deliver the prompt average multiplicity *Mγ(p)* or energies *Eγ(p)* and $\varepsilon_\gamma(p)$. If *S(E)* is a total γ-ray spectrum, then – total average values *Mγ(t)*, *Eγ(t)* and $\varepsilon_\gamma(t)$. It is obvious that average total multiplicity should be the sum of prompt and delayed ones: *Mγ(t) = Mγ(p) + Mγ(d)*.

The γ-ray spectrometers used in the experiments have the specific low energy cut-off or detector threshold (E_{thr}) as well as the high energy limit (E_{max}). The integration within these limits results in the partial multiplicity $M_\gamma(E_{thr} < E\gamma < E_{max})$ and average energy $E\gamma(E_{thr} < E\gamma < E_{max})$. Since the γ-ray energy spectrum from ^{252}Cf(s.f.) decreases exponentially above ≈ 1 - 2 MeV, the integration above 5 - 6 MeV has no practical sense because the variation of the integral becomes smaller than experimental uncertainties. It is not a case for the low energy limit E_{thr}. Therefore Table 4.2, additionally to the measured partial M_γ and E_γ, also lists the thresholds for every measurement.

Besides the integral quantities derived from the measured γ-ray energy spectra by authors, there are experiments where such quantities were measured directly or without reporting the gamma energy spectra. We found several such measurements for spontaneous fission of ^{252}Cf [75] - [83], which are listed in Table 4.2. There are publications which report very specific characteristics of γ-ray multiplicity:

dependence of $M\gamma$ on fragment charge ratio [84], standard deviations of multiplicity distribution [85], mixed prompt and delayed $M\gamma$ [86], dependence on the fission fragment total kinetic energy [87], etc. However, they do not provide the average multiplicities or energies – the quantities of interest for the present evaluation – and we do not see a way to derive them from the data measured in those experiments.

All collected experimental partial prompt and total γ-multiplicities are plotted in Fig. 4.6. versus the γ-ray detection threshold (low energy cut-off) E_{thr} used in the individual experiments. This plot definitely shows that partial total multiplicity $M_\gamma(t, E\gamma > E_{thr})$ is approximately two times larger than prompt $M_\gamma(p, E\gamma > E_{thr})$ and both monotonically decrease with the increase of the detection threshold (the eye-guided line highlights this tendency).

Fig. 4.7 displays the average partial γ-ray energy $<E\gamma>(E\gamma > E_{thr})$ released in spontaneous fission of ^{252}Cf in the prompt (p) and total (t) emission modes. As can be seen, $<E\gamma(t)>$ is about 2 times as much as $<E\gamma(p)>$. This is mainly due to the similar ratio between corresponding multiplicities $M\gamma(t)$ and $M\gamma(p)$. The spectral average energies ε_γ of PFGS and TFGS differ only by ≈ 10%. The slightly larger hardness of the total emission spectrum in comparison with the prompt one is explained by the energy distribution of the delayed γ-rays only up to ≈ 1.8 MeV.

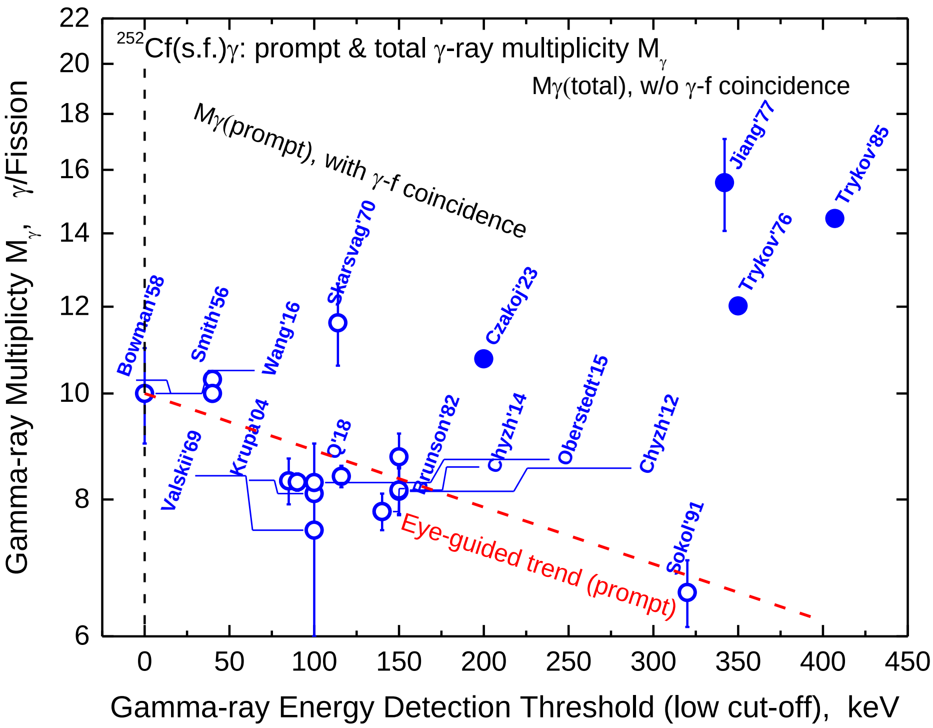

FIG. 4.6. Total and prompt partial γ-ray multiplicities for ^{252}Cf(s.f.) versus the detection threshold (low energy cut-off). The known measurements are shown by blue symbols: open – prompt gamma emission, closed – total emission. The red dashed curve is the eye-guided trend.

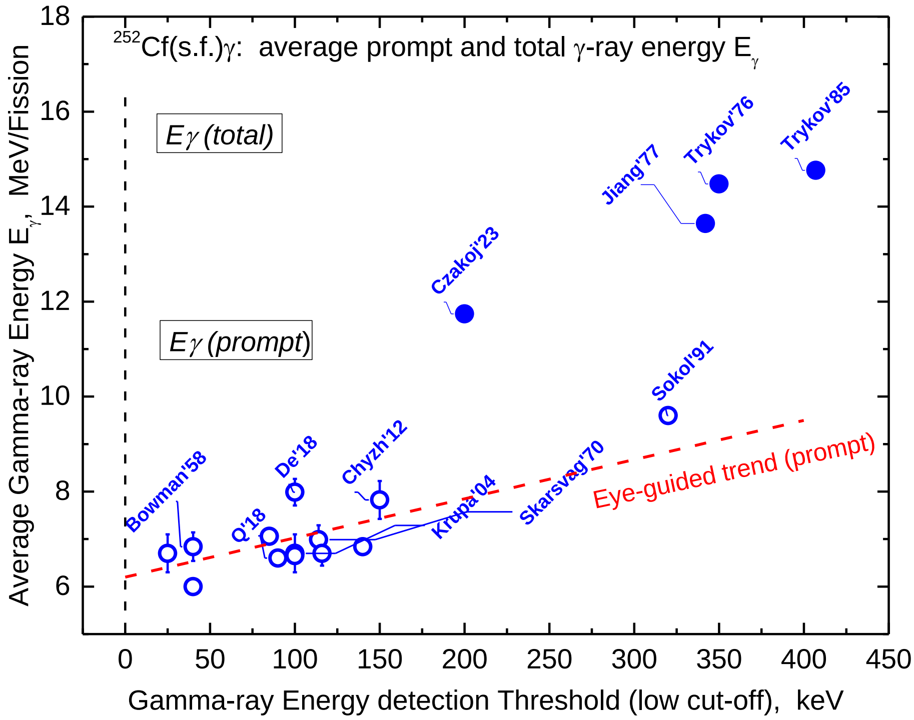

FIG.4.7. Total and prompt partial average γ-ray energy per spontaneous fission of ^{252}Cf versus the detection threshold (low energy cut-off). The known measurements are shown by blue symbols: open – prompt gamma emission, closed – total. The red dashed curve is the eye-guided trend.

5. GMA fit to the prompt ^{252}Cf(s.f.) γ-ray energy spectrum, average multiplicity and energy

5.1. The GMA code and its modifications

The non-model evaluation of the energy spectrum of the prompt γ-rays from ^{252}Cf(s.f.) was performed by means of the least-squares method implemented in the Gauss-Markov-Aitken code GMA (which was accompanied by the input data preparation code DAT) [4], [5]. For decades, these codes were used for the evaluation of the neutron induced cross section within the IAEA "Standards" project (see sub-directory "Codes and Programs" on the web-page [2]). During this period the original version [4], [5] has undergone several modifications, the latest versions being GMAP and DATP [2].

For the actual task, an evaluation of the ^{252}Cf(s.f.) prompt γ-ray energy spectrum, further modifications became necessary. To process numerous input data sets with many energy points we modified the GMAP code by increasing the length of the corresponding arrays. This was done by declaration of array lengths as parameters instead of fixed values. Then the values of the parameters were increased until the GMAP ran without crush.

The DATP code had the hard-wired interpolation "Log-Log" law below the neutron E_n (in our case – gamma E_γ) energy 0.03 MeV and "Lin-Lin" above this. The reason for this was an exponential increasing of the neutron cross sections as E_n decreases to thermal, but relatively flat cross sections in the fast neutron energy range. Since the ^{252}Cf(s.f.) prompt and total γ-ray spectra decrease exponentially versus the gamma energy E_γ, the Log-Log interpolation was declared also for $E_\gamma > 0.03$ MeV. Fig. 5.1 illustrates the impact of the interpolation law changes on the GMA evaluated PFGS. It can be seen that the Log-Log law results in a notable increase (in average by ≈ 10%) and smoothing of PFGS in the energy domain $E_\gamma > 5$ MeV. In particular, the spectrum fluctuations in the energy domain 8 - 15 MeV are smoothed out.

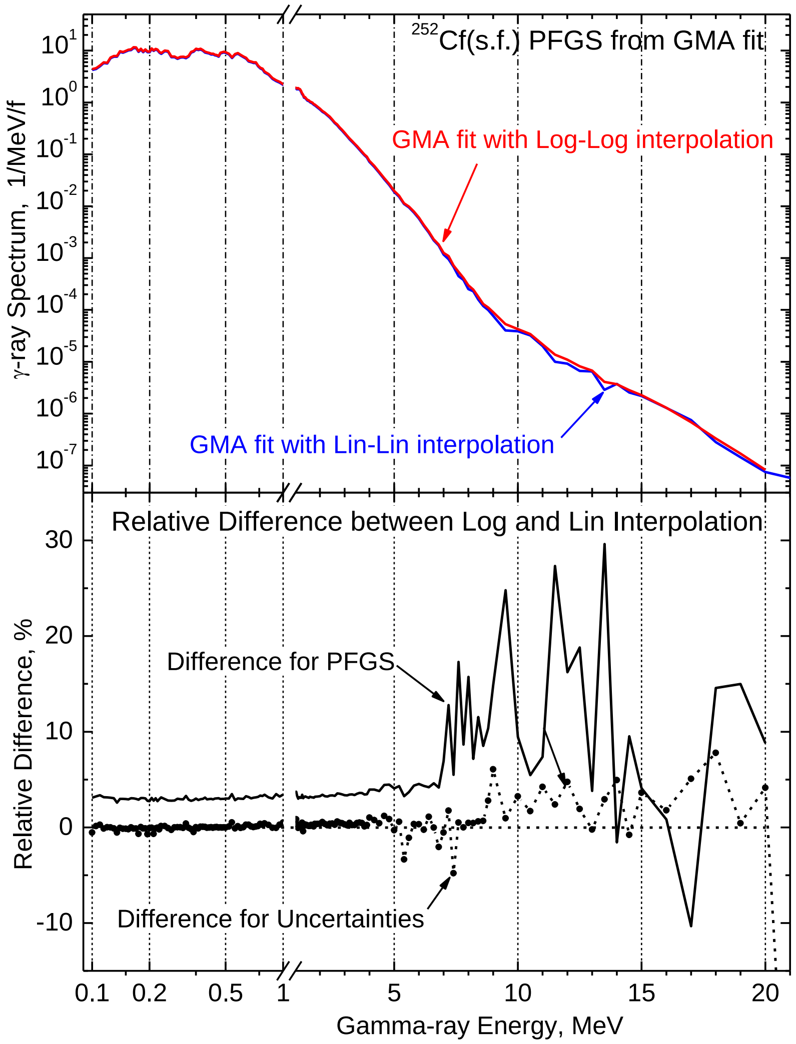

FIG. 5.1. Impact of the interpolation law change (from Lin-Lin to Log-Log) in the DATP code on the ^{252}Cf(s.f.) prompt fission gamma spectrum. Top: PFGS with Lin-Lin (red curve) and Log-Log (blue curve) interpolation laws. Bottom: the relative differences between evaluated spectra and uncertainties. Note the change of the γ-ray energy scale at 1 MeV.

The systematic up-shift (in other words, the "interpolation law bias") ≈3% is also observed at the lower gamma energies (but this bias is within the GMA evaluated uncertainty 2 - 3%, see Fig. 5.2). The implementation of Log-Log interpolation resulted in the overall chi-square parameter χ^2 decreasing from 3.94 to 2.74. The resultant GMA evaluated uncertainties for PFGS increased above 5 MeV by ≈3% in average.

Further modifications of the GMAP and DATP codes include the transition from programming language Fortran-77 to Fortran-95, which allowed for more flexible coding and compilation controls. The implementation of the free format for the input data, numerous comments and print-outs, make the input of data and reading of results much more user friendly.

5.2. Prompt fission gamma energy spectrum (PFGS)

Following the collection and analysis of the known measurements presented in Section 4, 18 measurements of the prompt γ-ray energy spectra [18], [20], [21], [22], [23], [25], [26], [27], [30], [31], [32], [33], [36], [37], [41], [43], [44], [46] (including one derived from spectra assigned to fission fragment mass [51]) were eventually selected for the GMA fit. In terms of the GMA code processing, they are called "data blocks" [4], [5]. Among them 3 experiments provided 2 - 4 γ-spectra measured at the same facility but with a modified set-up (different γ-ray detectors, different distance to the Cf source, etc.) [23], [37], [41], [43]. In the GMA terminology such spectra are defined as "data sets". The corresponding data sets of one block were considered in our evaluation as partially correlated. In several cases, the energy ranges of the original datasets were narrowed to exclude the large deviations which are usually observed near threshold or at the highest energy end (the criterium was a comparison with the bulk of all known data). The prompt γ-ray energy distribution measured in 5 experiments [20], [21], [27], [36] and [37] were considered in the present fit as a shape data, other 13 – as absolute PFG spectra normalized per one ^{252}Cf spontaneous fission event.

The total emission γ-ray energy spectra measured in 4 experiments [67], [68], [72], [74] were also added to the GMA evaluation, but only above 1.8 MeV - the highest γ-ray energy expected from the delayed mode of the ^{252}Cf spontaneous fission. All these total gamma spectra were considered as a shape data (i.e., the relative ones), since the TFGS absolute values are notably larger than PFGS.

The energy domain below 100 keV was excluded from the non-model evaluation, since the existing measurements are scarce, the reported gamma spectral data are incomplete or fully informative, see Section 4.4.

Above 20 MeV the energy spectrum of prompt γ-rays from spontaneous fission of ^{252}Cf is not yet experimentally well established. The first results of J. Kasagi et al. [25], who reported the γ-ray yield up to 160 MeV, were rather unexpected, so many other labs attempted to check his results. As shown in Fig. 4.3, all currently known γ-spectra measured in the energy range 20 to 160 MeV differ by 2 - 3 orders of magnitude. The comparison of PFGS for ^{252}Cf(s.f.)γ with the gamma production in other fission reactions could also be informative. Thus, Fig. 4.3 also displays the spectrum from ^{235}U(n_{th},xγ) measured by V. Varlachev et al. [88]. As seen, his results tend to the lowest values of the ^{252}Cf(s.f.) γ-spectrum observed by D. Pandit et al. [46] in the underground experiment or by N. Eremin et al. [33]. Regarding this contradictive information the γ-ray energy range above 20 MeV was excluded from the present GMA evaluation.

The energy grid, i.e., the nodes at which the evaluated PFGS spectra will be produced, has to fulfil several criteria. The grid should be rather dense to reproduce the specific structure observed in the measured PFGS. As can be seen in Figs 4.1 and 4.2, such a resonance-like structure becomes visible for Eγ below 0.7 – 0.8 MeV (the objective reason is the discrete energy radiation transitions between the first levels in the fission fragment nuclei). At higher energy the PFGS spectrum should be and indeed looks smoothed due to the statistical de-excitation mechanism of the dense nuclear levels.

The width of the observed quasi-discrete lines depends on the resolution of the used gamma detectors. The highest resolution was achieved with detectors which contained heavy elements, as for example BaF_2, $LaCl_3$ or $LaBr_3$ crystals in experiments [41], [43], [44]. Fig. 5.2 displays the energy resolution of the stilbene, Si(Li), NaI and $LaBr_3$ detectors, which we managed to find in the corresponding publications. It shows that the relative energy resolution varies from 3 to 20%, the minimal value is being reached for the $LaBr_3$ γ-detector. Fig. 5.2 also plots the selected relative increase of the grid $\Delta E_\gamma/E_\gamma$ versus γ-ray energy E_γ: it varies between 2 and 4% and is always smaller than the detector resolution. This is in particular true for the γ-energy domain below 1 MeV, where the experimentally observed quasi discreet structure has to be reproduced with sufficiently fine energy grid steps.

The energy interval between nodes ΔE_{grid} should also be rather small to keep the change of spectrum ΔPFGS from one point to another below a reasonable value. As can be seen in Fig. 5.2, below 1 MeV, where the PFGS in average is roughly flat, the relative oscillation of spectrum ΔPFGS / PFGS is within ≈ 20% which reflects the oscillating character of the spectrum itself. Beyond 2 - 3 MeV the spectrum is already smooth but starts to decrease exponentially, hence ΔPFGS / PFGS comes to be negative and increases up to -30% per energy step near $E_\gamma \approx 10$ MeV. Further decreasing ΔPFGS by reduction of the grid step ΔE_{grid} at this γ-ray energy is however not desirable from the view of scarce measured data and their increasing uncertainties.

For the final GMA evaluation in the gamma energy range 0.10 to 20.0 MeV a grid with 156 nodes was eventually selected, see Table 5.1. The measured points deviating from *a-priori* spectrum by more than three experimental uncertainty were excluded by pre-processing code DATP. The number of the fit iterations was restricted by 2, since the next one results to the spectrum changes which are lesser than the estimated uncertainty below gamma energy 8 MeV, but causes the visible artificial oscillations above.

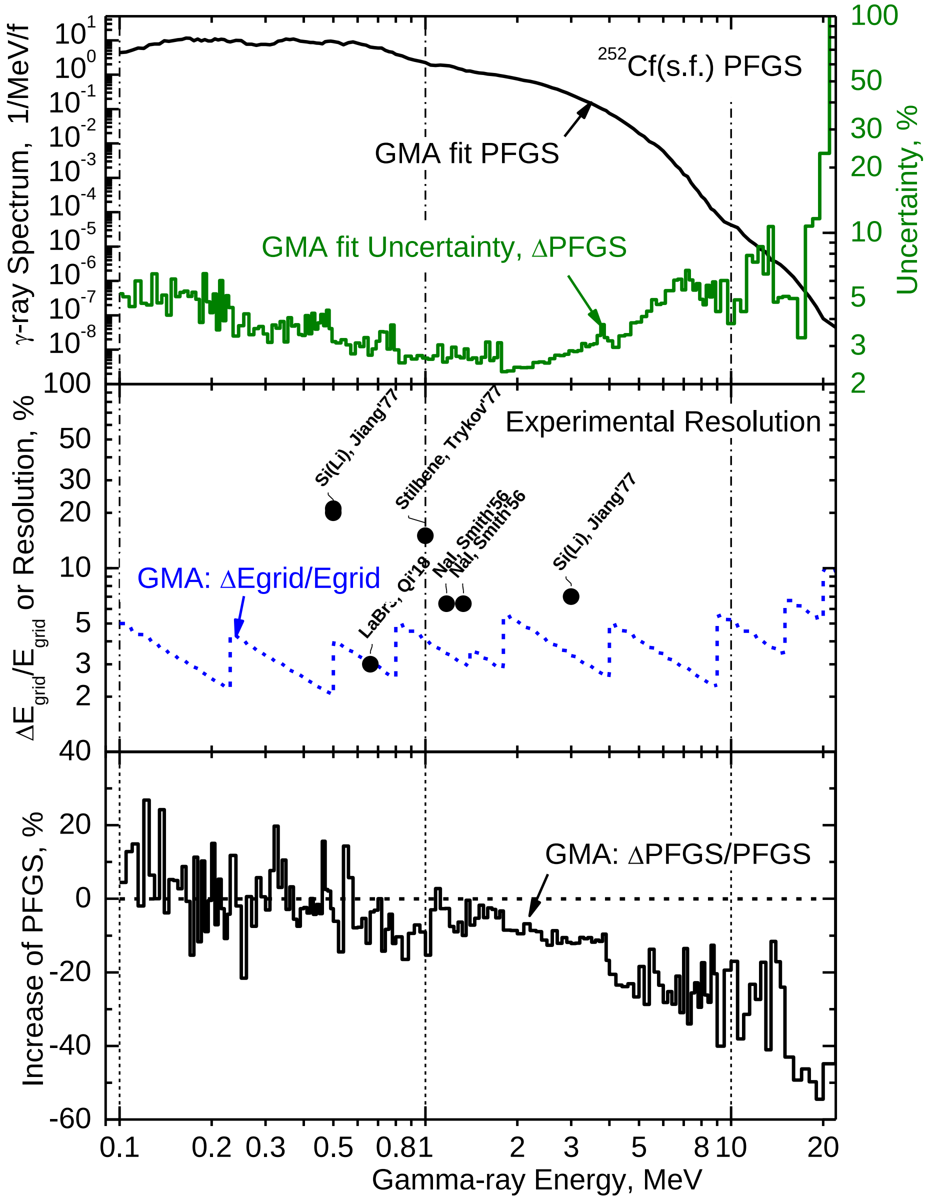

Fig. 5.2. ^{252}Cf(s.f.) PFGS and relative uncertainty evaluated by GMA versus the γ-ray energy (top); the variation of the relative energy grid step $\Delta E_{grid}/E_{grid}$ and energy resolution for several detectors and experiments (middle); relative change of the evaluated prompt spectrum ΔPFGS/PFGS from one grid point to another (bottom).

TABLE 5.1 Recommended prompt fission γ-ray spectrum and relative uncertainty for ^{252}Cf(s.f.) non-model evaluated in the present work by means of the GMA code.

Eγ, MeV	PFGS, γ/MeV/f	Rel. Unc., %	Eγ, MeV	PFGS, γ/MeV/f	Rel. Unc., %
0.100	4.367E+00	5.23	1.200	1.797E+00	2.64
0.105	4.561E+00	5.06	1.240	1.662E+00	2.96
0.110	5.147E+00	4.56	1.280	1.512E+00	2.69
0.115	5.915E+00	5.99	1.320	1.416E+00	2.65
0.120	5.799E+00	4.72	1.360	1.275E+00	2.91
0.125	7.356E+00	4.63	1.400	1.270E+00	2.60
0.130	7.828E+00	6.46	1.450	1.179E+00	2.64
0.135	7.830E+00	4.74	1.500	1.116E+00	2.50
0.140	9.725E+00	5.16	1.550	1.097E+00	2.66
0.145	9.354E+00	4.16	1.600	1.041E+00	3.13

Eγ, MeV	PFGS, γ/MeV/f	Rel. Unc., %	Eγ, MeV	PFGS, γ/MeV/f	Rel. Unc., %
0.150	9.845E+00	6.12	1.650	1.015E+00	2.57
0.155	1.034E+01	5.08	1.700	9.854E-01	2.65
0.160	1.062E+01	5.30	1.750	9.341E-01	3.11
0.165	1.155E+01	5.38	1.800	9.031E-01	2.28
0.170	1.146E+01	5.08	1.900	8.262E-01	2.30
0.175	9.708E+00	5.31	2.000	7.551E-01	2.39
0.180	1.081E+01	4.93	2.100	6.832E-01	2.38
0.185	9.543E+00	3.85	2.200	6.369E-01	2.39
0.190	1.052E+01	6.47	2.300	5.819E-01	2.52
0.195	9.581E+00	4.80	2.400	5.301E-01	2.53
0.200	9.541E+00	4.27	2.500	4.707E-01	2.50
0.205	1.098E+01	5.22	2.600	4.112E-01	2.62
0.210	1.021E+01	3.56	2.700	3.755E-01	2.72
0.215	1.075E+01	6.01	2.800	3.301E-01	2.71
0.220	1.046E+01	3.93	2.900	2.952E-01	2.75
0.225	9.329E+00	5.13	3.000	2.602E-01	2.85
0.230	8.936E+00	4.52	3.100	2.285E-01	2.83
0.240	9.988E+00	3.33	3.200	2.009E-01	2.86
0.250	9.793E+00	3.73	3.300	1.798E-01	3.08
0.260	7.678E+00	4.22	3.400	1.603E-01	2.98
0.270	7.723E+00	3.38	3.500	1.434E-01	3.04
0.280	7.144E+00	3.61	3.600	1.264E-01	3.08
0.290	7.556E+00	3.69	3.700	1.123E-01	3.35
0.300	7.596E+00	3.41	3.800	9.925E-02	3.76
0.310	7.365E+00	3.12	3.900	8.974E-02	3.28
0.320	7.930E+00	3.75	4.000	7.471E-02	3.17
0.330	9.497E+00	3.62	4.200	5.937E-02	2.96
0.340	9.789E+00	3.89	4.400	4.546E-02	3.34
0.350	1.082E+01	3.13	4.600	3.462E-02	3.38
0.360	1.051E+01	4.06	4.800	2.663E-02	3.83
0.370	1.085E+01	3.71	5.000	1.952E-02	3.87
0.380	1.025E+01	3.76	5.200	1.593E-02	4.11
0.390	9.486E+00	3.39	5.400	1.135E-02	4.35
0.400	9.230E+00	3.36	5.600	9.795E-03	4.91
0.410	9.009E+00	4.14	5.800	7.843E-03	4.67
0.420	8.661E+00	3.42	6.000	5.998E-03	4.73
0.430	8.669E+00	4.25	6.200	4.306E-03	5.41
0.440	8.298E+00	3.59	6.400	3.220E-03	5.41
0.450	8.172E+00	3.84	6.600	2.297E-03	6.05
0.460	7.841E+00	4.20	6.800	1.815E-03	6.10
0.470	9.066E+00	3.72	7.000	1.253E-03	5.45
0.480	9.293E+00	4.39	7.200	1.084E-03	6.70
0.490	9.486E+00	3.61	7.400	7.150E-04	6.06
0.500	9.229E+00	3.15	7.600	5.319E-04	5.43
0.520	8.661E+00	3.10	7.800	4.103E-04	5.83
0.540	7.409E+00	3.21	8.000	2.893E-04	4.93
0.560	8.471E+00	3.03	8.200	2.391E-04	4.65
0.580	8.957E+00	2.76	8.400	1.766E-04	5.72
0.600	8.250E+00	3.10	8.600	1.269E-04	5.05
0.620	7.611E+00	2.98	8.800	1.109E-04	5.93
0.640	7.204E+00	3.28	9.000	8.828E-05	4.33
0.660	6.330E+00	2.72	9.500	5.293E-05	6.02
0.680	6.104E+00	2.91	10.000	4.265E-05	3.81

Eγ, MeV	PFGS, γ/MeV/f	Rel. Unc., %	Eγ, MeV	PFGS, γ/MeV/f	Rel. Unc., %
0.700	5.916E+00	3.15	10.500	3.540E-05	4.90
0.720	5.917E+00	3.50	11.000	2.191E-05	4.33
0.740	5.075E+00	3.48	11.500	1.502E-05	7.86
0.760	4.654E+00	2.90	12.000	1.153E-05	7.32
0.780	4.459E+00	3.75	12.500	8.377E-06	8.63
0.800	3.915E+00	2.88	13.000	6.930E-06	6.45
0.840	3.510E+00	2.51	13.500	4.083E-06	10.69
0.880	2.932E+00	2.69	14.000	3.611E-06	4.78
0.920	2.657E+00	2.61	14.500	2.994E-06	5.02
0.960	2.468E+00	2.71	15.000	2.275E-06	5.08
1.000	2.245E+00	2.64	16.000	1.296E-06	4.97
1.040	1.901E+00	2.61	17.000	6.571E-07	3.27
1.080	1.844E+00	2.70	18.000	3.530E-07	10.72
1.120	1.896E+00	3.03	19.000	1.774E-07	11.62
1.160	1.845E+00	2.53	20.000	8.075E-08	23.29

The final GMA evaluation of the prompt γ-ray energy spectrum for spontaneous fission of ^{252}Cf is shown in Fig. 5.2 and listed in Table 5.1. In total, N_{tot} = 1435 experimental points in a measurement vector V and uncertainty U (after interpolation of all experimental γ-ray spectra to the 156 grid nodes) were included in the GMA fit to find the optimal solution F. The consistency between the experimental data discrepancies and assigned uncertainties was performed with the help of the reduced chi-square parameter per degree of freedom (or number of unknown parameters):

$$\chi^2 = \frac{1}{(N_{tot} - N_{adj})} \sum_{i=1}^{N_{tot}} \frac{(V_i - F_i)^2}{U_i^2} \qquad (5.1)\ ,$$

where N_{adj}= 167 is the number of the adjusted parameters of the fit, which is the sum of the number of the points in *a-priory* gamma spectrum N_{apr} = 156 and the amount of the shape spectra sets N_{shp} = 11.

The resultant GMA fit reduced chi-square parameter per degree of freedom χ^2 amounts to 2.8. The moderate exceeding of the unity means non-crucial underestimation of the uncertainties given in the original publications or those assumed by us if proper information was absent (as described for every experiment in Section 4). It also points to the possible underestimation of the uncertainties for the evaluated PFGS.

The contributions of the individual experiments (i.e., the data blocks and additionally the sets, if any) to the GMA fitting procedure are displayed in Fig. 5.3. The top half of Figure shows the number of points (after reduction to the grid nodes by DATP) from each experiment and the total number of points accumulated during sequential inclusion of each next experiment in the GMA fit. As can be seen, for example, the experiment of A. Oberstedt et al. (this block consists of 4 data sets) [41] adds the maximal number of points 534 to the totally accumulated in fit 1435.

The bottom part of Fig. 5.3 displays the reduced chi-square parameter χ^2 estimated for each experiment (data block) and χ^2 accumulated block-by-block in the GMA fit. The calculations were performed using Eq. (5.1) but the number of unknown parameters were either the number of points in block or number accumulated in the GMA fit, respectively. As can be seen, the shape data (labelled in Figure with letter S after the author of experiment in parenthesis; the number there is an amount of the sets in block) have as a rule smaller chi-square parameter than absolute γ-ray spectrum data. The reason is obviously the rescaling of the spectrum, which the GMA code automatically applies to the shape data. Fortunately, the recent and highest quality experiments have individual χ^2 close to unity. Consequently, the reduced chi-square parameter accumulated at the end of the GMA fit comes to relatively small value 2.6.

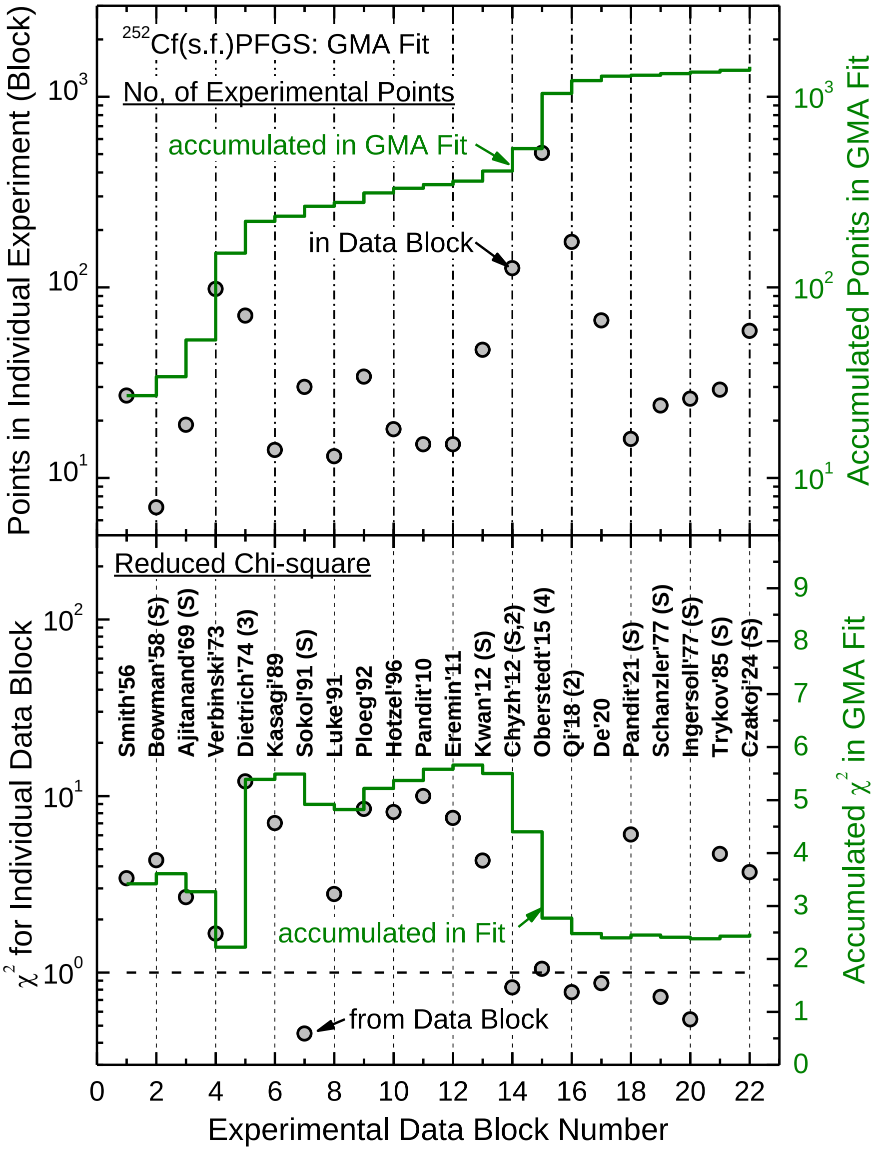

Fig. 5.3. Fitting of the ^{252}Cf(s.f.) PFGS. Top: number of points in the individual data block (circles) and accumulated in the GMA fit (histogram). Bottom: reduced chi-squared for the individual data block (circles) and accumulated in the GMA (histogram). Labels indicate the author and year of experiment (data block), in parenthesis the type of data (S - shape) and the number of sets in block are given.

The energy-energy correlation matrix obtained in the GMA fit is displayed in Fig. 5.4. As seen the moderate strength correlation up to ≈ 0.5 are observed in some energy domains. This reflects the correlations in the proper measurements which provided simultaneously 2 - 4 datasets with partially modified set-ups. In other Eγ - Eγ areas the evaluated PFGS for ^{252}Cf(s.f.) has no correlations. This results from the fact that all considered measurements did not use any common reference nuclear data. The key experimental parameters (such as the Cf-source strength, detector efficiency or response matrix, unfolding etc.) were independently measured and processed in each lab.

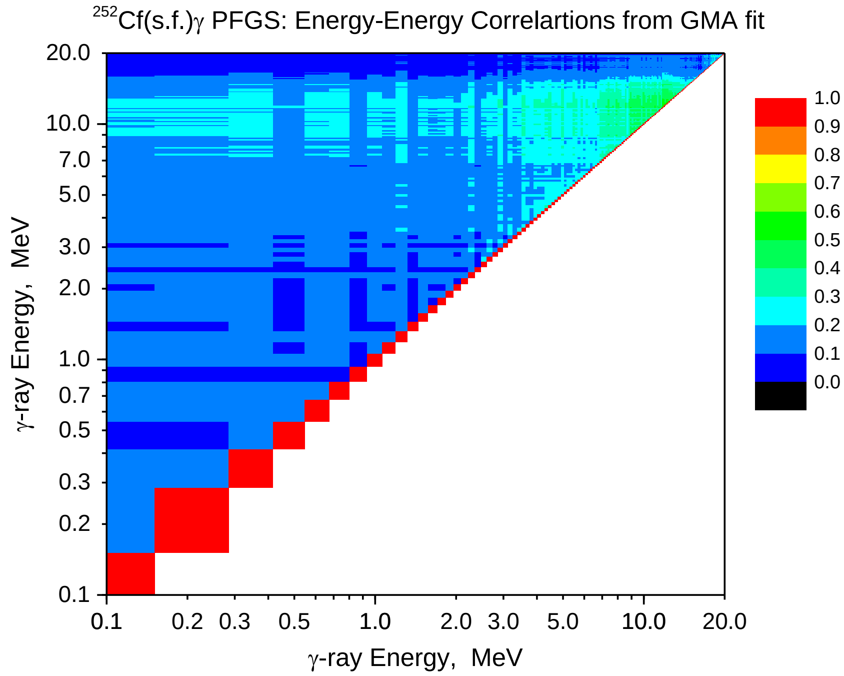

FIG. 5.4. Energy-energy correlation matrix for the $^{252}Cf(s.f.)$ PFGS evaluated by GMA in the γ-ray energy range 0.1 – 20 MeV.

5.3. The average multiplicities and γ-ray energies.

The GMA code [4], [5] allows to calculate additional quantity by integration over the energy spectrum given as independent input. In previous practice, i.e. in the evaluation of the neutron reaction cross section in frame of Standards [1], the prompt fission neutron spectrum from ^{252}Cf(s.f.), ^{235}Uf(n_{th},f) or other reference fields often served as such energy spectra. Then the independently measured neutron spectrum averaged cross sections (n-SACS) in those fields could be used as additional constraint for the non-model evaluation [1], [17].

Presently there is no known experiment where the ^{252}Cf(s.f.) prompt or total gamma fields were used to measure the gamma induced spectrum averaged cross sections (γ-SACS). The same is true for any other γ-ray energy continuous field, since there was no large interest in such data. Consequently, additional experimental γ-SACS data cannot be used in the GMA fitting procedure. Instead, in the present PFGS evaluation, the artificial flat γ-ray distribution was used to integrate the evaluated prompt fission γ-ray spectrum and its correlation matrix with the help of GMA. By doing so the partial prompt multiplicity within the energy range 0.10 to 20 MeV and its uncertainties could be calculated. However, we do not use this quantity as independent parameter in the GMA fitting procedure since most of Mγ were obtained by integration of the measured γ-ray energy spectra which are already part of our evaluation.

The partial prompt multiplicity $M\gamma(p, E_\gamma > 0.1\ MeV)$ was estimated by integration of evaluated PFGS with the help of GMA and equals 8.158 γ/f. When the low energy integration limit (threshold or energy-cut off) E_{thr} varies, then this results in the dependence of partial $M\gamma$ on E_{thr} graphically shown in Fig. 5.5. As can be seen, the partial multiplicity $M\gamma(p, E_\gamma > E_{thr})$ calculated with evaluated PFGS reasonably reproduces the known experimental data measured with different energy thresholds.

As shown in Section 4.4, the scarce and incomplete existing experimental data did not allow an estimation of the prompt gamma spectrum below 100 keV. However as it was shown, the spectrum evaluated by D. Stoddard reasonably reproduce the measured data at least in the energy interval 5 – 50 keV. For the estimation of the prompt γ-ray multiplicity in the whole energy range we integrated Stoddard' PFGS from 0 to 100 keV which brought 1.036 photons/f. This value was added to $M\gamma(p, E_\gamma > 0.1\ MeV)$ = 8.158 γ/f to get the prompt multiplicity integrated in the entire energy range: $M_\gamma(p, E_\gamma > 0\ MeV)$ = 8.158 + 1.036 = 9.194 γ/f (or 2.440 γ/ν_n, after division by total neutron multiplicity ν_n(t) = 3.7676 n/f). As can be seen in Fig. 5.5, such an interpolation to zero threshold visually agrees with the observed tendency of the $M\gamma$ dependence on the low energy cut-off parameter.

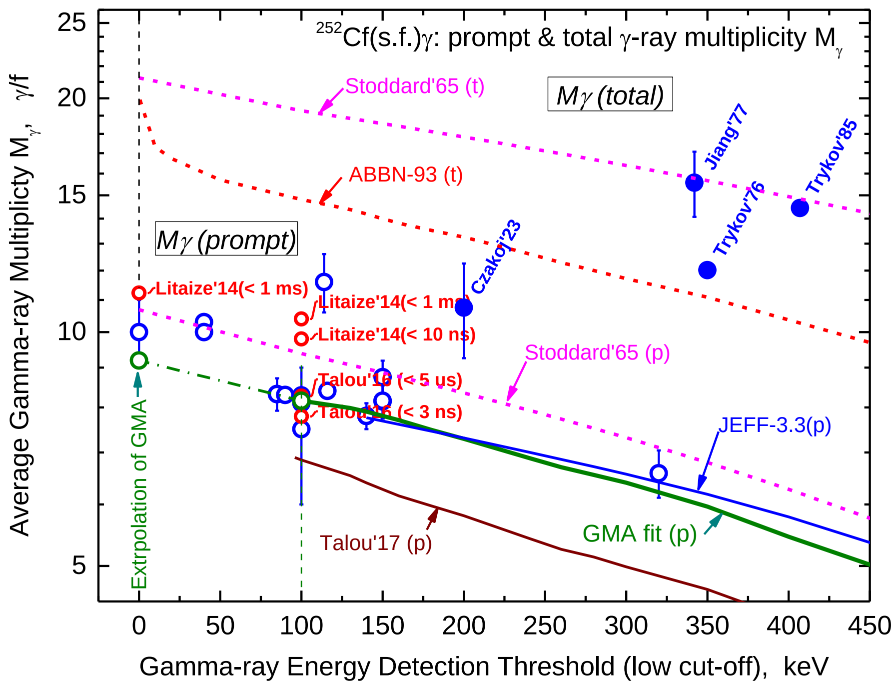

FIG. 5.5. Total and prompt γ-ray multiplicities for ^{252}Cf(s.f.) versus the low-energy detection threshold. The known measurements are shown by blue symbols: open – prompt emission, closed – total. GMA evaluation of the present work - solid green curve; extrapolation to zero threshold – green dash-point curve. Theory: red symbols (FF disintegration time is given in parenthesis) from [89], [90], [91]; brown curve from [91]. Existing evaluations: solid and dashed curves with corresponding reference.

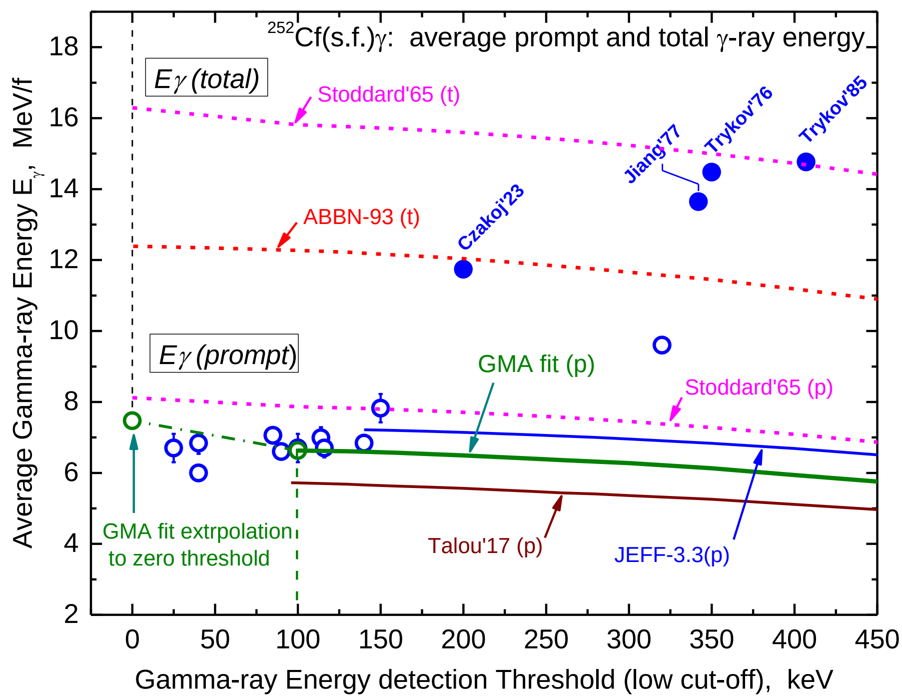

FIG. 5.6. Total and prompt average γ-ray energy for the ^{252}Cf(s.f.) source versus the detection threshold. The known measurements are shown by blue symbols: open – prompt emission, closed – total. GMA evaluation of the present work: solid green curve; extrapolation to zero threshold - green dash-point curve. Existing evaluations: solid and dashed curves.

Fig. 5.6 displays the average prompt gamma energy released per fission $E_\gamma(p)$ versus the energy cut-off parameter. The partially integrated prompt average energy $E_\gamma(p, E_\gamma > cut\text{-}off)$ agrees with the majority of experimental data. Extrapolation to zero energy brings average gamma energy per fission $E_\gamma(p, E_\gamma > 0\ MeV)$ = 7.472 MeV/f or the spectrum averaged energy $\varepsilon_\gamma(p, E_\gamma > 0\ MeV)$ = 0.813 MeV (after division by $M_\gamma(p, E_\gamma > 0\ MeV)$ = 9.194 γ/f).

6. Comparison of reference ^{252}Cf(s.f.) prompt γ ray energy spectrum with theory and major evaluated libraries

Theory. For the energy of γ-rays below ≈ 10 MeV, the prompt γ emission accompanying the ^{252}Cf spontaneous fission originates from the statistical de-excitation cascades and from the decay of giant dipole resonances in the excited fission fragments. Theoretical studies were undertaken in many publications including those where the measured PNGS were reported (see publications in section 4). As an example of the most recent theoretical investigations we refer to the works of O. Litaize et al. [89] and P. Talou et al. [90]. They have shown that for Eγ < 12 MeV the main mechanism is statistical radiation de-excitation of the excited states in the fission fragments. The used statistical reaction models which depend in-turn on several parameters and nuclear structure: yields of the fission fragments, giant dipole resonance (GDR) parameters, excited level density models, etc.

In the energy domain Eγ ≈ (12 - 190) MeV the emission mechanism is a bremsstrahlung during the fission fragment acceleration in the Coulomb field. The proper models use the classic or quantum approaches, their free parameters still have to be fitted to experimental data. As was shown in Section 4.2, the available experimental data demonstrate a spread of several orders of magnitude at the γ ray energies above ≈ 20 MeV. The prompt gamma spectrum calculated by S. Maydanyuk et al. in 2010 [92] is somewhere in between the experimental data known before this year, see Fig. 4.3. However, the recent and likely most reliable underground measurement of D. Pandit et al. in 2022 [46] results in a PFGS which is a 100 times smaller than the one predicted by S. Maydanyuk et al. [92]. Both the experimental and theoretical information about the ^{252}Cf(s.f.) prompt γ-ray spectrum above 20 MeV is far from an acceptable quality.

Comparison of the theoretical calculations of P. Talou et al. [91] (available in energy range 96 keV - 9.86 MeV) with PFGS evaluated by GMA in the present work is shown in Fig. 6.1, 6.2 and 6.3 for the γ-ray energy ranges 0.07 – 1.2 MeV, 1 – 10 and 5 – 20 MeV, respectively. As can be seen, the theoretical predictions of P. Talou et al. [91] disagree with the GMA fit up to factor 2 in the considered energy range. It has to be noted that theoretical calculations slightly depend on the time interval (typically nano – milli seconds) during which the gamma emission is modelled [89], [90]. The calculation of S. Maydanyuk et al. [92], available from 5 MeV and 20 MeV, generally reproduces the overall trend of the non-model evaluated PFGS but deviates at some energies by a factor up to 10, see Fig. 6.3.

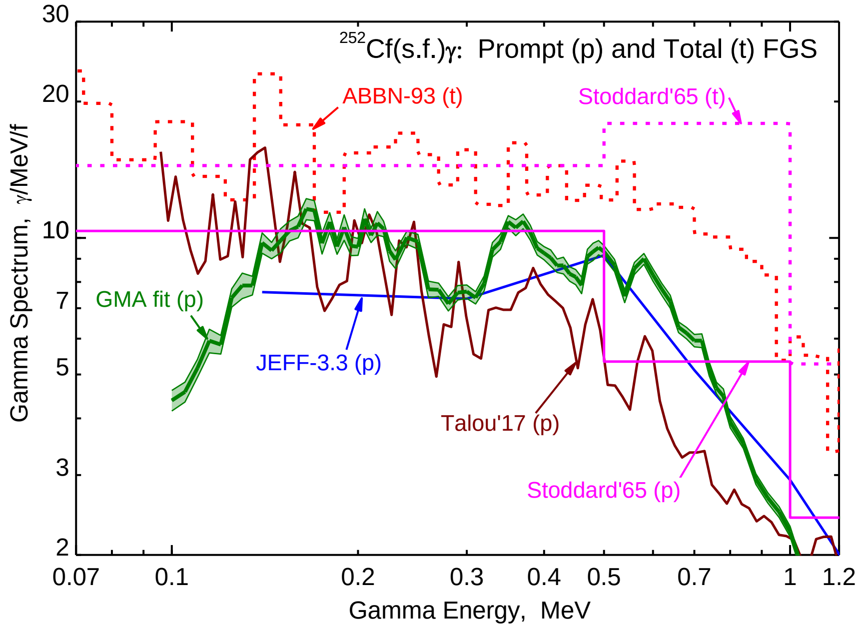

FIG. 6.1. Prompt (p) and total (t) γ-ray spectra from ^{252}Cf(s.f.) in the energy range 0.07 – 1.2 MeV. GMA evaluation of the present work - prompt γ-ray spectrum (green curve with uncertainties). Theory - prompt γ-ray spectrum calculated by P. Talou [90], [91] (brown curve). Existing evaluations - prompt and total γ-ray spectra of D. Stoddard [13], [14] (solid and dashed pink histograms); total γ-ray spectrum from ABBN-93 [72] (dashed red histogram), JEFF-3.3 [8] – prompt (blue curve).

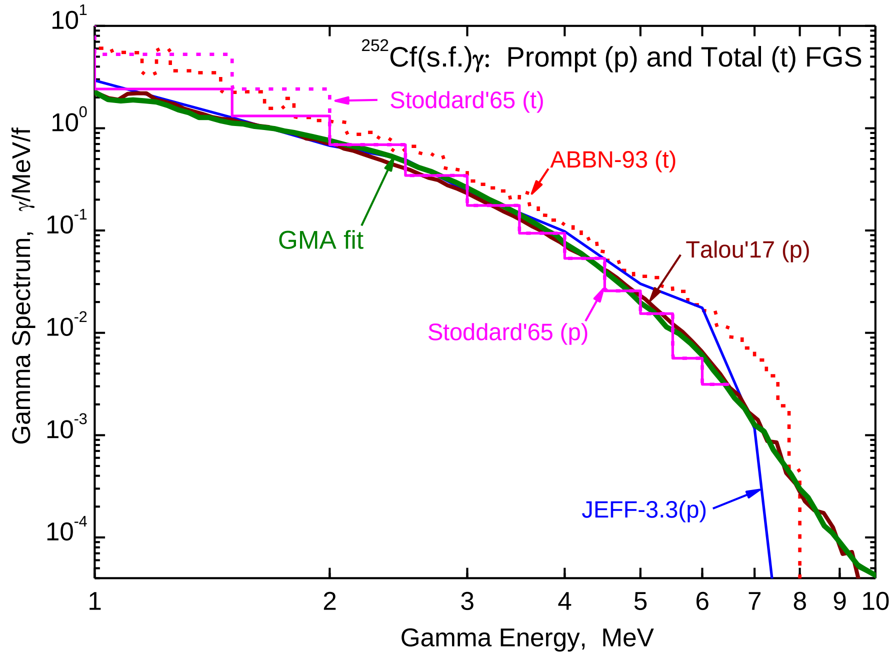

FIG. 6.2. Prompt (p) and total (t) γ-ray spectra from ^{252}Cf(s.f.) in the energy range 1.0 – 10 MeV. GMA evaluation of the present work - prompt γ-ray spectrum (green curve with uncertainty). Theory - prompt γ-ray spectrum calculated by P. Talou [90], [91] (brown curve). Existing evaluations - prompt and total γ-ray spectra of D. Stoddard [13], [14] (solid and dashed pink histogram); total spectrum from ABBN-93 [72] (dashed red histogram), JEFF-3.3 [8] – prompt (blue curve).

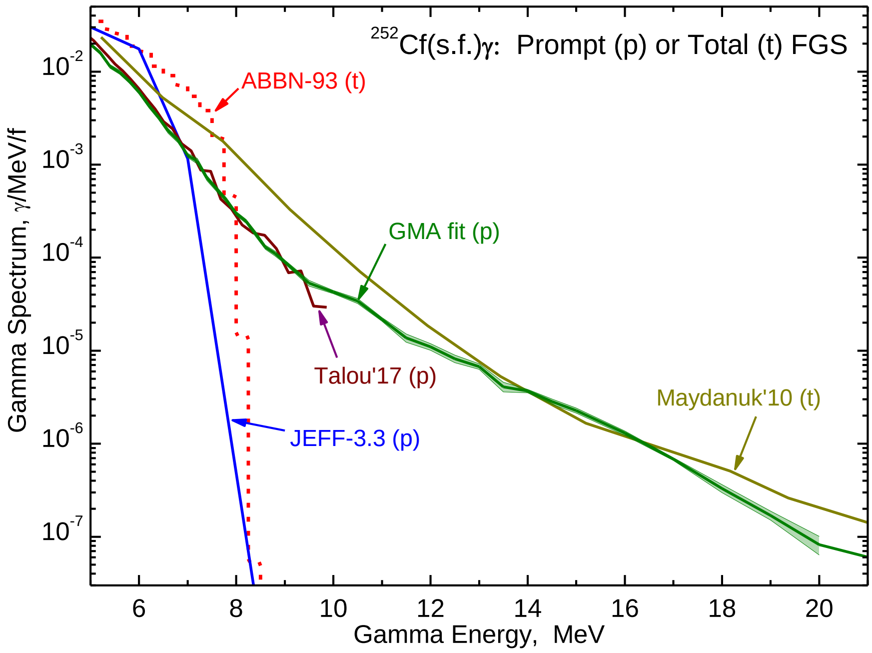

FIG. 6.3. Prompt (p) and total (t) γ-ray spectra from ^{252}Cf(s.f.) in the energy range 5 – 22 MeV. GMA evaluation of the present work - prompt γ-ray spectrum (green curve with uncertainty). Theory - prompt γ-ray spectrum calculated by P. Talou [90], [91] (brown curve), total γ-ray spectrum of S. Maydanyuk [92] (dark yellow curve). Existing evaluations - total γ-ray spectrum from ABBN-93 [72] (dashed red histogram), JEFF-3.3 [8] – prompt (blue curve).

Evaluations. The status of the prompt, delayed and total γ-ray energy spectra was analysed in the following libraries.

(1) US Evaluated Neutron Data Files ENDF/B-VIII.0 [7]. The following data for photon emission from isotope ^{252}Cf are presented in the Radioactive Decay Data sub-library NSUB = 4, as material MAT = 3644, in the file MF = 8 and section MT = 457:

- 3 discrete γ-lines with energies Eγ = 43,4, 100.2, 160.0 keV, which are the radiation deexcitation transitions in the nucleus ^{248}Cm produced by α-decay of ^{252}Cf;
- 5 X-ray lines with energies Ex = 19,6 – 126.6 keV and their intensities, which are the radiation deexcitations in the electron shell of the Cm atom.

It is important to stress that ENDF/B-VIII.0 has no continuous γ-spectra from spontaneous fission of ^{252}Cf, neither prompt, nor delayed nor total.

(2) Joint European Fission and Fusion file JEFF-3.3 [8]. Data for three processes of photon emission from ^{252}Cf are represented as material MAT = 3675, in the file MF = 8 and section MT = 457:

- 3 discrete γ-lines with energies Eγ = 43.4, 100.2 and 160.0 keV, which are the radiation deexcitation transitions in the nucleus ^{248}Cm produced by α-decay of ^{252}Cf (similar to ENDF/B-VIII.0, but different intensities);
- continuous γ-spectrum in the energy range 140 keV to 10 MeV with comment *"Gamma spectrum = Verbinski et al. Phys. Rev. C7(1973)1173*";
- 7 X-ray lines with energies Ex = 5,0 – 127.0 keV, i.e., the XLαβγ, XKαβ rays, which are the radiation deexcitations in the electron shell of the Cm atom after α-decay of ^{252}Cf (similar to ENDF/B-VIII.0, but different energies and intensities).

It is worthwhile to underline that the continuous γ-spectrum presented in JEFF-3.3 is the spectrum accepted from the single experiment: V. Verbinski et al. [22] published in 1973. The total or delayed γ-ray energy spectra are not available in this library.

(3) The Russian evaluated data library ABBN-93 [93]. Regrettably we did not find the information how the ^{252}Cf(s.f.) γ-ray energy spectra were evaluated or where it originates from. The numerical values of

the total γ-ray energy spectrum (referenced here as ABBN-93) was provided in the database ICSBEP, Entry Alarm-Cf-Fe-Shield-001 [72]. There it was recommended to use given spectrum for simulation of the gamma radiation transport in the iron spheres with ^{252}Cf source. The spectrum is tabulated as γ-ray yields per one Cf-neutron in 127 groups from 50 keV to 10.8 MeV.

(4) Besides the complete evaluated data libraries, the prompt, delayed and total γ-ray spectra for ^{252}Cf(s.f.) were found in reports of D. Stoddard published in 1965 [13], [14]. The reports summarized the Cf radiation properties known in the literature and probably calculated by the author himself. The numerical energy spectra are given there for the prompt gammas in the energy range 0.0 to 6.5 MeV, delayed – 0.0 to 2.0 MeV and total – as a sum of PFGS and DFGS. The data are presented in the energy groups which have the rather large width 0.5 MeV.

The prompt or total γ-ray energy spectra from these four evaluations are co-plotted together with PFGS from the present GMA fit in Fig. 6.1 – 6.3. JEFF-3.3, which adopted the prompt γ-ray energy spectrum from the single experiment (see (2) above), suffers from obvious drawbacks: scarcity of points below 1 MeV and incapability to reproduce the structure there; substantial disagreement at higher energies and too low high-energy limit ≈ 8 MeV. ABBN-93, which provides only the total γ-ray energy spectrum, visibly overestimates our GMA PFGS above ≈ 1.8 MeV, where however both PFGS and TFGS have to coincide from a physical point of view. At high energies the ABBN-93 spectrum is limited by 9 MeV. D. Stoddard provided a rather coarse energy bin (0.5 MeV) for the PFGS spectrum below 1 MeV, but still reasonably reproduces the GMA fit there in average (Fig. 6.1). It also reasonably agrees at energies beyond 2 MeV, but Stoddard's evaluation ends at 6.5 MeV.

The total and delayed γ-spectra from spontaneous fission of ^{252}Cf are not presented in the JEFF-3.3 and ENDF/B-VIII.0 evaluations.

Gamma multiplicity and average energy derived from the major evaluated libraries. These integrated values were calculated from the prompt and total gamma energy spectra presented in the considered evaluated libraries. Their dependencies on the variable low energy integration limit (threshold or cut-off) up to 0.450 MeV are displayed in Figs 5.5 and 5.6.

The prompt experimental energy integrated quantities, i.e., the prompt average multiplicity Mγ(p) and energy Eγ(p), are reasonably reproduced by the JEFF-3.3 and Stoddard evaluations (excluding the drawback of latter – too wide energy bin 0.5 MeV).

The energy integrated quantities for total gamma emission, Mγ(t) and Eγ(t), are measured in a small number of experiments and the results are broadly scattered, see Figs 5.5 and 5.6. The proper data from ABBN-93 and D. Stoddard differ by ≈ 20% from measured data and among themselves. The integrated and energy differential characteristics of the total γ-ray emission from ^{252}Cf(s.f.) were not evaluated in the present work.

7. Delayed γ-ray spectrum, average multiplicity and energy from spontaneous fission of ^{252}Cf: existing measurements and evaluations

The key distinguished parameter of the delayed gammas accompanying the fissioning nuclei is the time interval after the prompt radiation emission. This time period is defined by the physical processes and spans of half-lives from ≈ 3 ns to several days. Of especial importance is a time moment separating the prompt and delayed radiations. This time boundary obviously has some uncertainty, since the measured radiation time decay curve consists of a prompt peak and a tail composed of several exponents with $T_{1/2}$ ≈ 10, 30 and 100 ns [57], [58], [60] (see more details in Section 4.4 and in this Section). Following the practice in the referenced publications we accepted the value 3 ns and consider the emission of photon radiation after this time as delayed one. The main mechanisms contributing to the delayed γ-ray emission are the de-excitation of: (i) the longer living states in the post-neutron-emission primary fission fragments (FF), (ii) the long-lived isomers in the primary FF and (iii) the excited states in the daughter nuclei produced by the β decay of the fission fragments. The first two processes are the pre-betta-decay emissions. Internal electron conversion of the γ-rays with energy below ≈ 100 keV may result in the suppression of these gammas and consequent emission of X-rays, which however follows immediately after.

The yields of (quasi) discrete transitions or smoothed energy spectra of the delayed fission gamma spectra (DFGS) from spontaneous fission of ^{252}Cf were rather seldom measured. We found publications of four teams: S. Johansson from the University of California [94], W. John et al. from Lawrence Livermore National Laboratory [95], R. Clark et al. from Argonne National Laboratory [96] and E. Reber, R. Gehrke with co-workers from Idaho National Laboratory [97], [98]. They used different experimental techniques, but still, in three of them the discrete γ-ray transitions (in contrast to the quasi-smooth energy distributions of the prompt radiation) were observed. The measured yields include the γ-rays from the de-excitation of the excited nuclei states and K X rays from the internal electron conversion associated with these gamma-rays. The highest energy end of delayed γ-rays found in these experiments does not exceed 1.8 MeV. The energies of delayed X rays from ^{252}Cf(s.f.) are smaller than ≈ 50 keV.

The next part of this section gives an overview of these experiments and their results relevant to the purpose of the present study. The principal parameters of these measurements are summarised in Table 7.1.

(1) In 1964, S. Johansson reported the measurements of the delayed gamma radiation emitted in the time range 10 - 300 ns from fission fragments of ^{252}Cf [94]. A very strong Cf-source (≈ 10^7 fissions per minute) was placed at one end of an evacuated tube, at the other end was a fission counter. A heavy lead shield was placed in such a way that the gamma radiation and neutrons from the source could not reach the scintillation spectrometer. This configuration allowed recording the radiation emitted only after the fragments travelled about 10 cm. Hence the delayed radiation could be studied without interference from the prompt radiation.

The fission fragments were detected by two semiconductor detectors which gave the pulses proportional to the kinetic energy of the fragments [94]. Since the mass ratio is inversely proportional to the ratio of the kinetic energies of two fragments, their masses could be determined from the observed pulse-height ratio of two detectors. The selection of the different mass ratios allowed the authors to investigate the gamma radiation as a function of mass.

The γ-rays were detected by a 5.0 cm × 7.5 cm sodium iodide crystal placed at 90° relative to the flight-direction of the fragments which minimized the neutron background [94]. The distance between the source and the scintillation spectrometer was 10 cm. This was meant to give good detection efficiency and, at the same time, avoid disturbing summing effects from coincident γ-rays. Most of the delayed radiation was emitted after the fragments had already reached the fission counters which were placed at about 1 cm distance from the source. This meant that the two fragments of a fission event were well separated at the time of emission and made it possible to study the radiation from only one fragment by placing a lead absorber over the other fission counter.

The main correction to the γ-ray spectra made by the authors was the subtraction of the background caused by neutrons [94]. A sodium iodide crystal has an efficient sensitivity to fast neutrons and to neutrons scattered in the surroundings which however take a longer time to reach the spectrometer. Correction for accidental coincidences was in general of little importance.

The time decay curve for all fragments was measured, it exhibited a prompt peak and a tail [94]. The shape of the tail showed several components with different half-lives. In the range 20 - 30 ns after fission, the half-life was of the order of 15 ns and in the period 200 - 300 ns later it had increased to about 100 ns. The partial decay curves were measured for the mass ratio intervals 1.0 - 1.2, 1.2 - 1.48 and > 1.48. All these partial decay curves have a pronounced tail, but the shape and mean half-life were different. The curve for R = 1.2 - 1.5 had the shortest half-life $T_{1/2}$ = 32 ns, for R > 1.5 the mean $T_{1/2}$ was ≈ 40 ns, the curve for R = 1.0 - 1.2 was complex with a half-life increasing to about 100 ns.

The experimenters determined the intensity of the delayed radiation in different ways, but they all reached nearly the same result [94]. The largest uncertainty was introduced by the fact that the shape of the decay curve was not known for times shorter than 10 ns and longer than 200 ns after fission. The necessary extrapolations were performed employing the $T_{1/2}$ = 15 ns of the decay for times shortly after fission and 100 ns for times longer than 200 ns after fission. The average value for the intensity of the delayed radiation relative to the prompt radiation was found to be 6.5% which corresponds to 0.65 photons per fission [94].

The authors of the experiment [94] studied the energy spectra of the delayed radiation for a few selected mass regions. The neutron background was determined in a separate experiment and subtracted. The low-energy peak in the spectra, 95 keV, was identified with the transition from the 2^+ state in the fragments with A ≈ 152. The energy of the 2^+ state for A ≈ 110 should then be 140 keV. The experimental value was 130 keV which was considered satisfactory agreement. The high-energy distribution extends up to 1.6 MeV which is a very reasonable energy value for the first 2^+ state in magic nuclei.

The energy spectra plotted in Fig. 5 of the Johansson' paper [94] are the pulse height distributions of the gamma counts in the energy range from 0.05 to 1.8 MeV for three numbers of fragment masses A = 132, 152 and 110. Obviously, this type of experimental data can neither be used for our analysis nor for comparison with other measured or evaluated data. The single information from this experiment, which could be relevant for the present study, is the experimenters' estimate of the delayed radiation multiplicity 0.65 photons/fission assigned, however to the specific fission fragments.

(2) In 1970, W. John with co-workers reported the results of measurement of γ-rays emitted during the first 3 - 2000 ns after spontaneous fission of ^{252}Cf [95].

The ^{252}Cf source was deposited onto 90 µg/cm^2 Ni foil and had a strength of 46 µCi [95]. It was positioned between two Si diodes which detected the oppositely directed fragments from a fission event. These detectors were cooled to -40^{o}C in order to keep the leakage current low as radiation damage accumulated. The resolution for fission fragments (peak-to-valley ratio) was between 2.8 and 2.9. A 9 cm^3 planar Ge(Li) crystal, located near one of the Si detectors, detected γ-rays emitted by fragments stopped on that Si detector. A 10 µm Be window separated the Ge crystal from the vacuum chamber. The energy resolution (FWHM) was 2 keV at 122 keV and 4 keV at 1.33 MeV.

A collimator of tungsten alloy shielded the Ge detector from the Cf-source, it also defined the fragment emitting the γ-rays and eliminated most of the prompt γ-rays [95]. A fast coincidence within 30 ns was required between the two fission pulses. The time-of-flight (TOF) measurement was started on the fission-fragment signal and stopped on the γ-ray signal from the Ge detector. Pile-ups of the more than one fast fission pulses were excluded within 2 µs which prevented the coincidence between the numerous α-particles and the fission fragments. The accidental time coincidence rate at level 17% was subtracted from the TOF spectrum. The timing resolution (FWHM) for γ rays increased from 2 ns at 1 MeV to ≈10 ns at 100 keV. The energy discriminator on the γ-ray was set at 80 keV, and the spanned γ-ray energy region was up to 3 MeV.

The physical quantities derived by experimenters in their work [95] included the post-neutron-emission fission fragment (FF) mass (M) distributions and the delayed γ-ray spectra delayed in the time interval 50 to 2000 ns and sorted by the FF masses. The total γ -ray spectrum was quite complex, so the events were sorted according to mass and into three major time intervals. The γ-ray peaks were well resolved, a computer search for γ-ray peaks was made by fitting the spectra with Fourier series. The counts in the window were then summed to obtain a three-dimensional array of counts versus the γ-ray energy E and mass M. The completed fit to the data peak yielded the number of counts, the gamma energy, fragment mass and the statistical standard deviations of these quantities. For each γ-ray the intensity per fission and the intensity per fission fragment were calculated from the number of counts under the peak and from half-life.

The authors of this work [95] showed that, while delayed γ-rays occur for most masses, the photon intensity is concentrated on certain masses, notably 91, 96, 99, 108 and 110 on the light-mass peak plus 134, 137 and 146 on the heavy-mass peak. Practically no delayed γ-rays come from the minima near masses 102 and 139 in spite of the high yields of these fragments. W. John et al. also pointed out that the data of S. Johansson [49] for prompt γ-ray yields show dips at the corresponding masses which was particularly evident at masses 108 and 134. The correlation between prompt and delayed yields suggests, according to [95], that the occurrence of delayed transitions simply subtracts radiation from the prompt yield, i.e., that the total yield as a function of mass is a relatively smooth sawtooth curve.

W. John et al. [95] listed the results of their data analysis in tabulated form. For each γ-ray (to be uncertain by ± 0.2 keV) and the most probable mass M (uncertainty ≈ 4.5 mass units) the intensities are given in terms of photons per spontaneous fission with standard deviation. The half-life is also given for each γ-ray. For half-lives less than 10 ns only strong or isolated lines were analyzed because of the large background from prompt γ-rays. Below 80 keV only the strongest lines were analyzed. Between 70 and 80 keV no lines were analyzed, because of interference from the lead X-rays. The experimenters estimated that the total γ-ray energy from unresolved lines is at most 20% of that of the resolved lines for $200 < E < 2000$ keV and $t < 100$ ns.

We used 144 discrete γ-rays emitted during the first 3 - 2000 ns after spontaneous fission of ^{252}Cf which are given in Table I of W. John et al. [95] (the authors seem not to have identified any one transition as X-rays). We folded the corresponding yields, i.e., the number of photons per fission, with the energy resolution having the Gaussian shape and relative energy resolution ΔFWHM / FWHM = 3%. The obtained energy spectrum of the delayed γ-rays is shown in Fig. 7.1.

(3) In 1972, R. Clark and co-workers measured the γ-rays and associated K X rays from the isomeric decay of fission fragments, populated in spontaneous fission of ^{252}Cf, but before the beta decay [96]. A thin source of ^{252}Cf (intensity 2 10^5 f/min) was placed between two Si-Au surface barrier (SB) detectors which detected the complementary fragment pairs from fission events. The apparatus was placed in a 9 inch diameter by 5 inch deep vacuum chamber maintained at a pressure of 1 mTorr of Hg.

A Si(Li) detector for the K X rays and low-energy γ-rays was positioned to be in full view of the face of each fragment detector, but shielded by a 1.7 mm thick Cu shield from prompt radiations emitted near the source position [96]. A Ge(Li) detector for higher energy photons was placed on the opposite side of the source from the Si(Li) detector. This detector viewed fragments stopped on the face of fragment detector F1 but was shielded by a Cu-lined Pb shield from prompt radiations emitted near the source and from delayed radiations from the fragment detector F2. Thus, photons were observed directly only if emitted by fragments stopped on a fragment detector face or by fragments in the last portion of their flight path.

The experimenters [96] determined the energy dependent absolute detection efficiency of X rays for the Si(Li) detector by placing standardized sources of ^{65}Zn, ^{75}Se, ^{93m}Nb, ^{109}Cd, ^{125}I, ^{133}Ba, ^{159}Gd and ^{241}Am on the face of a used fragment detector. The response functions for these K X ray standards were in good agreement with published values, using the energies and relative intensities of the $K_{\alpha1}$, $K_{\alpha2}$, $K_{\beta1}$ and $K_{\beta2}$ components. The energy resolution, full width at half maximum (FWHM), for the 26.36 keV γ-ray of ^{241}Am was 560 eV. The energy-dependent absolute detection efficiency and linearity of the Ge(Li) photon detector were determined similarly but with sources ^{241}Am, ^{109}Cd, ^{57}Co, ^{203}Hg, ^{137}Cs and ^{60}Co. The energy resolution (FWHM) of this detector was 2.0 keV for the 122.1 keV transition of ^{57}Co and 2.9 keV for the 1332 keV transition of ^{60}Co.

The electronics used in the experiment [96] blocked any event in which two pulses from two fragment detectors occurred in the time interval of 20 to 10 ns. This reduced the distortion of the fragment kinetic energy spectrum caused by accidental pile-ups between fission fragments and alpha particles. A pulse shape discriminator circuit was used to reject Si(Li) pulses with anomalous rise times. The signal from the Ge(Li) detector was set to trigger on pulse heights corresponding to 25 keV gamma rays.

The coincidence logic of signals from two fission fragment detectors, F1 and F2, and from the Si detector of X-rays or from the Ge(Li) detector of γ-rays allowed to record three types of events: F1-F2-X, Fl-F2-gamma or F1-F2-γ-X. From these events the photon energy spectra were accumulated, which were sorted into time-after-fission 8 intervals (5 to 3000 ns) for Ge(Li), 10 intervals (7 to 3000 ns) for Si(Li) and 2 amu wide mass intervals for the SB detector. Each mass-time sorted photon energy spectrum was computer analysed to locate peaks and intensity. The elemental identifications were made on the basis of the γ-X coincidence, similar mass and half-life assignments, also including information known before [95].

R. Clark and co-workers presented the measured isomeric photon intensities per ^{252}Cf(s.f.) in two tables: those assigned to the specific nuclei and without such specification [96]. The maximum energy of γ-rays was reported to be 1.338 MeV, the half-lives from ≈ 2 to 4100 ns. *We used 130 assigned (after excluding 10 lines with half-lives $T_{1/2} \leq 3$ ns) and 105 unassigned X and gamma transitions from these two tables in [96]. The discrete X- and γ-ray yields were folded with Gaussian distribution having relative energy resolution ΔFWHM / FWHM = 2% (an average between 2.0keV / 122keV and 2.9keV / 1332keV). The obtained spectrum is plotted in Fig. 7.1.*

(4) In 2004, E. Reber, R. Gehrke and co-workers published their first results [97]. The first experiment used a ~ 10 year old source of ^{252}Cf. Due to this reason several observed γ-rays were attributed to the disintegration of other isotopes of californium: ^{249}Cf with half-life $T_{1/2}$ = 351 y and ^{251}Cf with $T_{1/2}$ = 898 y. In 2005 this group reported new measurements with a newly prepared (~ 1 year old) ^{252}Cf source with activity 50 µCi [98]. The overall size of the capsule is approximately 1 cm in diameter and 0.5 cm thick with a 0.25 mm thick beryllium foil window on one side and a backing of 0.127 cm thick stainless steel on the other side. The californium was embedded in an anodized aluminium disk and sealed in the capsule.

Gamma-ray spectra from the Cf sources were acquired with a large coaxial Ge detector [98]. This detector was calibrated with calibrated sources at many γ-ray energies ranging from 121.7 to 2614.5 keV. An uncertainty of ±5% was assigned to the Ge detector efficiency between 121 and 1400 keV and ±8% from 1400 to 2614 keV.

The experimenters managed to detect hundreds of peaks among innumerable fission product γ-rays from the spontaneously fissioning ^{252}Cf [98]. The identification and quantification of specific fission products was difficult because of the close spacing of the associated γ-rays and the large number of interferences. However, they solved this problem with the precise measurement of the γ-ray energies with uncertainties of < 0.1 keV and comparison with the γ-ray energies reported in the Evaluated Nuclear Structure Data File (ENSDF) with uncertainty ± (0.2 - 0.3) keV. The majority of γ-ray emission probabilities, Pγ(E), for the more intense fission product γ-rays were also known to < 5% in ENSDF. Also, the evaluated fission yield compilation of England and Rider in 1994 allowed to indicate which fission products are primarily cumulative, Y_c, which are independent, Y_i, and which are a combination of both. The accuracy of these parameters permitted the authors an unique identification of many fission-product γ-ray peaks as well as the dominant component when the multiple radionuclide assignments were possible (radionuclides for which $Y_i > Y_c$ or $Y_c \times P_\gamma(E)$ is greatest for a peak of energy E) [98]. While many of the γ-ray peaks below 1 MeV have been uniquely identified, quantitative measurement of their yields was difficult due to multiple fission product assignments which occurred more frequently at these lower energies. From comparison of these measured γ-rays per spontaneous fission with the independent FF yield the authors concluded that in most observed fission product γ-rays the independent yield is a small fraction of the total measured yield.

The measured γ-lines from fissioning ^{252}Cf(s.f.) and assignment to the producing nuclei are listed in Table 3 of paper E. Reber et al. [98]. The maximum discrete energy is 1.791 MeV. *We used the γ-ray energies and yields (number of rays per spontaneous fission) from this Table. To have only delayed gammas, the γ-ray transitions which were denoted by experimenters as prompt, as well as those directly populated in fission, were excluded. The selected 182 discrete lines (half-life from 0.92 s to 12.75 d) were then folded with the γ-detector energy resolution ≈3%. The obtained energy spectrum of the delayed gammas (DFGS) is plotted in Fig. 7.1.*

The energy spectra of the ^{252}Cf(s.f.) delayed fission γ-rays (DFGS) obtained by folding of the discrete data of W. John et al. [95], R. Clark [96] and E. Reber et al. [98] are intercompared in Fig. 7.1. It can be seen here that these spectra essentially differ from each other. The single point where they do agree is the production of the 1.13 MeV gamma quasi-discrete line, which is a superposition of the 1.103, 1.132 and 1.136 MeV γ-rays from deexcitation of ^{102}Tc, ^{135}I and ^{134}I, respectively [98]. At other γ-ray energies above 0.2 MeV the spectrum of E. Reber et al., measured in 2005, is substantially larger than that of W. John et al. [95] or that of R. Clark measured 20 – 25 years earlier. Most probably, the advanced technique used by E. Reber et al. allowed him to detect and identify a larger number of the discrete delayed γ-rays accompanying ^{252}Cf(s.f.).

The most prominent γ-ray transitions observed in DFGS, Fig. 7.1, result from the decay of the isomeric states in the fission fragments or from the daughter nuclei after β^- decay. For example, the 6^+ isomers in the even Z isotopes (fission fragments) ^{134}Tc and ^{136}Xe have half-lives 163 ns and 2.8 µs, respectively [96]. The de-excitation cascade $6^+ \rightarrow 4^+ \rightarrow 2^+ \rightarrow 0^+$ produces 1279 keV and 1313 keV gammas at the last stages.

The comparison of the these measurements with the single evaluation of DFGS by D. Stoddard [13], [14] indicates roughly agreeing results, but only in the first energy interval 0.0 - 0.5 MeV. In all other energy bins 0.5 - 1.0 MeV, 1.0 – 1.5 MeV and 1.5 - 2.0 MeV the evaluation of Stoddard substantially overestimates the presently existing experimental data. However, from our view, this disagreement points to the missing of numerous weak photon transitions in these experiments.

The theoretical calculations of DFGS for ^{252}Cf(s.f.) seem to be not available.

We also attempted to extract the delayed spectrum as a difference between the total (TFGS) measured by L. Trykov [70] - [72] and PFGS evaluated by GMA in the present work. The resultant spectrum for γ-ray energies of more than 0.4 MeV is depicted in Fig. 7.1. As can be seen it is larger than all known directly measured DFGS, but on the other hand supports the evaluation of D. Stoddard.

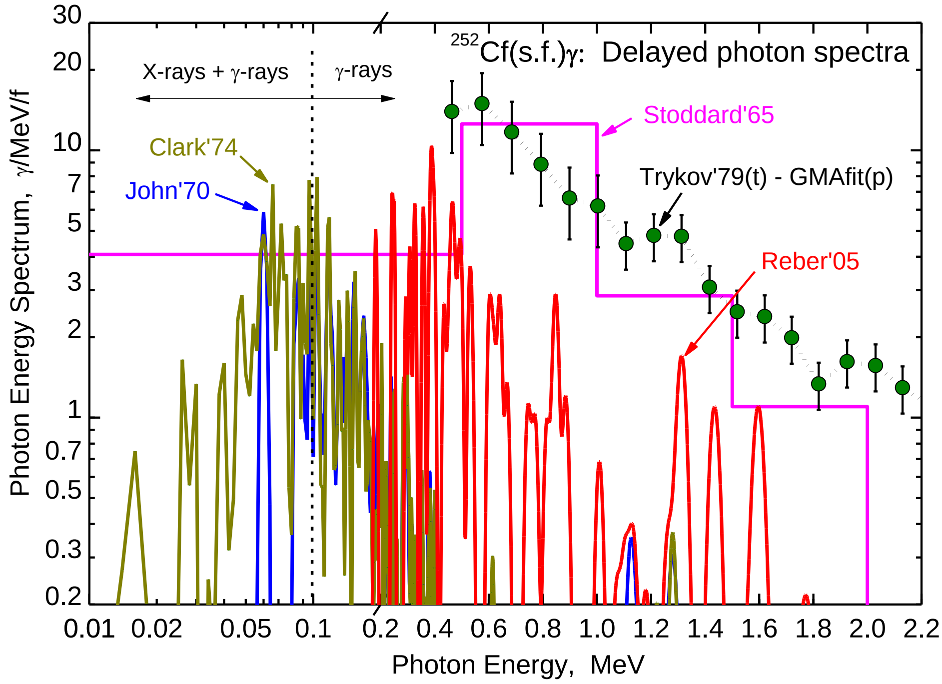

FIG. 7.1. Delayed X- and γ-ray spectra from ^{252}Cf(s.f.) in the energy range 10 keV – 2.1 MeV. Experimental discrete γ-yields (after folding with spectrometer energy resolution): W. John et al. [95] (blue curve), R. Clark et al. [96] (dark yellow) and E. Reber et al. [98] (red). The solid green symbols denote the delayed spectrum obtained by subtraction from the TFGS measured by L. Trykov [70] - [72] of PFGS evaluated by GMA in the present work. The single existing evaluation: delayed spectra of D. Stoddard [13], [14] (solid pink histogram). Note the change of energy scale at 0.2 MeV.

The average delayed γ-ray multiplicity Mγ(d) and energy Eγ(d) were extremely seldom measured or evaluated, see Table 7.2.

S. Johansson was the single author who directly reported the measured delayed multiplicity as 0.65 γ/f [94]. His value corresponds to the energy range of the recorded γ-rays between 0.05 and 1.80 MeV and emission time 10 to 300 ns. It is important to note that the author derived this value from integration of the pulse-height distributions (PHD), i.e., not from the γ-ray spectrum after proper unfolding. The other two known experiments [95], [98] did not reported the average multiplicity Mγ(d) or energy Eγ(d). We derived the partial gamma multiplicity Mγ(d) = 0.34 γ/f and energy Eγ(d) = 0.27 MeV/f from the experiment of W. John [95] by summing of the discrete yields within the energy interval [0.10 - 1.28] MeV. Similarly, Mγ(d) = 1.58 γ/f and Eγ(d) = 0.65 MeV/f were obtained from data of E. Reber et al. [98] in the interval E = [0.19 - 1.80] MeV. It has to be added that measured or assigned delay emission time ranges were substantially different in the W. John and E. Reber experiments: 3 ns – 2000 ns and 1 s – 13 days, respectively.

TABLE 7.1. The known measurements of the delayed discrete photons or their energy spectra from ^{252}Cf(s,f.): first author and laboratory, key parameters of experiments, references. The time interval during which the γ-rays were registered after the fission event is given in parentheses.

First Author	Year Lab	X- or γ-rays energy range, MeV	^{252}Cf strength, f/s	γ-ray detector	Fission Fragment (FF) detector	Time (coincidence window) and geometry selection of delayed photons	Corrections, Comments	Reference EXFOR Entry
S. Johansson	1965 LLNL	0.05 – 1.60	1.7E+05	Ø5.0×7.5 cm NaI(Tl)	Si semi-conductor	γ-FF coincidence (10 – 300 ns), shielding from prompt gammas	only PHD reported	[94]
W. John	1970 LLNL	0.08 – 3.00	1.7E+06	9 cm^3 planar Ge(Li)	two Si diodes	γ; γ-FF coinc. (3 – 2000 ns), shielding from prompt gammas	corrected discrete γ-rays	[95] 14479.002
R. Clark	1974 ANL	0.01 – 0.10 0.03 – 1.50	3.3E+03	S(Li) for X-ray Ge(Li) for γ	two Si-Au surface barrier detectors	γ-FF, γ-X-FF coinc. (5 – 3000 ns), shielding from prompt γ	corrected discr. K X and γ-rays	[96]
E. Reber	2005 INL, Idaho	0.10 – 1.79	1.9E+06	large coaxial Ge detector	no FF detection	γ-spectroscopy (≈ 1 μs)	corrected discrete γ-rays	[98]

TABLE 7.2. Measured or derived average delayed γ-ray multiplicity Mγ(d) per fission and spectrum averaged gamma energy ε_γ(d) for ^{252}Cf(s.f.)γ. The γ-ray energy range (threshold and high-energy limit) and emission time interval are quoted.

First Author, Lab (Year)	γ-ray energy range: threshold - high-end	γ-ray emission time range	Mγ(d), γ/f	ε_γ(d), MeV	Reference
Experiment					
S. Johansson, LLNL (1965)	50 – 1600 keV	10 – 300 ns	0.65	n/a	[94]
W. John, LNLL (1970)	80 – 1280 keV	3 – 2000 ns	0.34 [1)]	0.27 [1)]	[95]
R. Clark, ANL (1974)	10 – 1500 keV	5 – 3000 ns	0.51 [1)]	0.21 [1)]	[96]
E. Reber, INL (2005)	100 – 1600 keV	1 s – 13 d	1.58 [1)]	0.65 [1)]	[98]
Evaluation					
D. Stoddard, SRL (1965)	0 – 2000 keV	all delayed	10.29 [1)]	0.77 [1)]	[13], [14]

Note: 1) These Mγ(d) and ε_γ(d) were calculated in the present work by summing of the discrete yields reported in the original works or weighting them with the γ-ray energies.

It can be seen that the measured average emission quantities are substantially different and crucially depend on the experimental conditions. Obviously, these experiments provide only the partial multiplicity or average energy since the γ-ray energy and time emission ranges were restricted by the used detectors and measuring techniques. An additional reason is that only the strongest (not all) discrete transitions were measured or identified in these experiments.

The single evaluation of the delayed γ-ray spectrum was carried out by D. Stoddard in the rather old work published in 1965 [13], [14]. The DFGS is represented there for γ-ray energy range 0.0 to 2.0 MeV but within too coarse bins 0.5 MeV. As seen in Fig. 7.1, such binning does not allow to reproduce the energy structure observed in the measurements. The energy integration of the D. Stoddard' spectrum brings multiplicity Mγ(d) = 10.3 γ/f, whereas the weighting with the γ-ray energy – the spectrum averaged energy ε_γ(d) = 0.77 MeV, see also Table 7.2.

The highest experimental value for the delayed γ-ray multiplicity Mγ(d) = 1.58 γ/f was derived from data of E. Reber et al. [98]. This value amounts only to ≈ 15% of the total delayed multiplicity predicted by the evaluation of D. Stoddard. The average energy of delayed γ-rays per spontaneous fission of ^{252}Cf ε_γ(d) = 0.65 MeV measured by E. Reber turns out to be in reasonable agreement with the 0.77 MeV evaluated by D. Stoddard, however the data of W. John is 3 times smaller.

Summarising this section, it should be stated that delayed γ-ray emission accompanying spontaneous fission of ^{252}Cf has hardly been studied, neither experimentally nor theoretically. This is true for the gamma energy spectrum, average energy and emission multiplicity. As a consequence, the delayed ^{252}Cf(s.f.)γ data are not represented in the major evaluated cross section libraries. It is evident that further experimental and theoretical studies are necessary. Meantime, from our view, there is no other choice as to use the old evaluation of D. Stoddard for the analysis and simulation of applications when such information is required.

8. Gammas following the pionic radioactivity of ^{252}Cf(s.f.)

This section considers the possible contribution of the 68 MeV gammas to the ^{252}Cf(s.f.) total γ-ray emission spectrum. These gammas are created by the pionic radioactivity of Cf with subsequent π^0 decay into two photons.

In 1986, D. Ion, M. Ivascu and R. Ion-Mihai were the first who suggested that natural pionic and muonic radioactivity might occur as a branch of spontaneous fission [99] (additionally, see [100], [101]). In the fission of heavy elements an energy up to Q ≈ 200 MeV is available, which exceeds the mass of pions or muons and is thus sufficient to create light unflavoured mesons. The Bucharest group combined the energetics with considerations of phase space for 3-body final states and concluded that the branching ratio for pion and muon emission relative to spontaneous fission for ^{252}Cf should be $\Gamma_{\pi+\mu}/\Gamma_{sf} \approx 5\ 10^{-4}$ [100].

What is currently known about masses of neutral and charged pions or muons and their decay characteristics is the following [102]:

- neutral pion π^0: mass = (134.9768 ± 0.0005) MeV, mean life time τ = (8.43 ± 0.13)×10^{-17} s, dominant decay mode is to two gammas with fraction $\Gamma_{2\gamma}$ / Γ = (98.823 ± 0.034) %;
- charged pions $\pi^\pm$: mass = (139.57039 ± 0.0005) MeV, mean life time τ = (2.6033 ± 0.0005)×10^{-8} s dominant decay mode is to charged muon $\mu^\pm$ and muon neutrino ν_μ with fraction $\Gamma_{\mu\nu}$ / Γ = (99.98770 ± 0.00004) %;
- charged muons $\mu^\pm$: mass = (105.6583755 ± 0.0000023) MeV, mean life time τ = (2.1969811 ± 0.0000022)×10^{-6} s; dominant decay mode is to positron e^+ or electron e^-, electron antineutrino $\bar{\upsilon}_e$ and muon neutrino ν_μ with fraction $\Gamma_{e\bar{\upsilon}\nu}$ / Γ ≈ 100 %.

Energetically, neutral and charged pions or muons could be produced in fission, but the $\mu^\pm$ and π^0 have a lighter masses than $\pi^\pm$, thus $\mu^\pm$ and π^0 radioactivity can be expected with larger probability as was supposed in the first studies [99], [100].

The neutral pions decay with 98.85% probability into two γ-rays on a time scale of ≈ 1 10^{-17} s or practically immediately after birth. The kinetic energy of the π^0 can extend theoretically from 0 to 50 MeV. However, most probably it will be close to zero since the Coulomb field at the scission point forces the release of most of the Q value as kinetic energy of the fragments. The pair of gammas resulting from decay of zero energy pions will fly away at an angle close 180 degrees (back-to-back). The energy of each gamma is expected to be at least around half of the π^0 mass, or ≈ 67.5 MeV.

Thus, the neutral pions spontaneously emitted by the fissioning nucleus from its ground state could be an additional source of gammas. These γ-rays could impact on the high energy (E ≥ 30 MeV) part of the total or prompt γ-ray spectrum for ^{252}Cf(s.f.) considered in the present report. In opposite to π^0, the main decay modes of the charged pions and muons are not accompanied by electromagnetic radiation [102], hence their emission seems to have no impact on the γ-ray spectrum from ^{252}Cf(s.f.).

After the theoretical work of Ion et al. [99], the experimental search for neutral pionic nuclear radioactivity from spontaneous fission of Californium was undertaken in several laboratories in the time from 1987 to 1992 [103] - [107], see upper part of Table 8.1. The search for the neutral pionic radioactivity was experimentally carried out by the detection of the two high energy (≈ 68 MeV) gammas emitted at 180 degrees, employing conventional NaI scintillation detectors, lead-glass Cherenkov detector telescopes or more complicated spectrometers. As we see, the single principal difference from conventional measurements of PFGS or TFGS from ^{252}Cf(s.f.) is the two back-to-back oriented gamma detectors working in a time coincidence regime. Due to importance of these results for topic of present report these experiments are reviewed below.

J. Beene et al. [103] used a ^{252}Cf source deposited on the plate in a fast fission-ionization chamber. An additional plate with ^{239}Pu was placed in the ionization chamber to indicate the fission activity induced from cosmic rays. The γ-rays were detected by an array of 63 NaI(T1) crystals that covered a large fraction of the total solid angle. The pairs of high energy (> 10 MeV) gammas were selected in prompt time coincidence (1.2 ns). The energies and the correlation angle between the γ-rays were measured. The events during 100 ns after the fission signal from the ionization chamber, either from ^{252}Cf or ^{239}Pu, were recorded for analysis. The authors eventually deduced that the Cf spontaneous fission decay associated with π^0 emission probability < 1 10^{-9} of the normal spontaneous fission at a 90% confidence level [103].

C. Cerruti carried out an experiment at the Saclay laboratory in order to determine the ratio of neutral pion emission relative to the spontaneous fission of a ^{252}Cf source [104]. The source intensity was 10^5 fissions/s. Two collinear blocks of lead glass (15×15×35 cm^3) were used to count the gamma rays from neutral pion decay. The effective solid angle was 15% of 4π. The response, the efficiency and the resolution of these detectors were studied before. In order to reduce the background due to cosmic rays, the lead glasses were surrounded by several veto scintillators and lead glasses. With such an active shielding, the number of background events was reduced by a factor of one hundred. The authors obtained an upper limit for ratio $\Gamma_{\pi 0}/\Gamma_{sf}$ < 5 10^{-9} [104].

The same group of experimenters led by J. Julien and co-workers, performed a new measurement in an underground laboratory (Laboratoire Souterrain de Modane) [105]. In this experiment six lead glasses (15×15×30 cm^3) were used to detect γ rays issued from neutral pion decay. In this set-up, three combinations corresponded to a back-to-back emission of two 68 MeV gamma rays. Two additional lead glasses were used to measure the background and random coincidences with other detectors. An upper limit value of Γ_π/Γ_{sf} less than 10^{-12} was assigned by the authors at 95% confidential level [105].

S. Stanislaus et al. carried out a search for the spontaneous emission of neutral pions from Pb, Bi, Th, U and Cf [106]. They used two collinear large NaI(T1) detectors working within a 15 ns coincidence window. The main γ-detectors were placed in a large cosmic ray shield constructed of 1.2 cm thick plastic. This reduced the background by a factor of 35. The used a Cf source produced 1.2 10^7 n/s. To reduce neutron loading, the neutron shielding (about 15 cm bricks of borax) was located between the source and the NaI crystals. A correction of about 30% for the attenuation of the γ rays from any π events was applied. For ^{252}Cf an upper limit $\Gamma_{\pi 0}/\Gamma_{sf}$ < 3.3 10^{-10} with a 90% confidence limit was obtained in this experiment [106].

J. Knudson et al. measured the Cf neutral pion activity by the coincident detection of two photons emitted in the decay [107]. The source consisted of ≈ 10.9 μg of ^{252}Cf in a 0.32 cm thick steel capsulate and was surrounded by 5% boron-loaded polyethylene to attenuate neutrons. The gamma spectrometer had two arms consisting of three lead-glass planes converting the γ-rays into a charged-particle shower;

multiwire proportional chambers and thin plastic scintillation detectors served as timing counters. The tracks in the proportional chambers were analysed to determine the conversion vertex. The laboratory angle between the two photons, which is related to the total energy of the original π^0, was reconstructed from the vertices. The total energy was also obtained from summing the energy deposited in the converter planes, scintillators, and lead-glass calorimeters in which the showers were stopped. The authors of the experiment estimated an upper limit to the branching ratio for neutral pions emitted from spontaneous fission to be $\Gamma_{\pi 0}/\Gamma_{sf} < 1.37\ 10^{-11}$ with a 90% confidence level [107].

TABLE 8.1. Known measurements of the pion and muon radioactive decay accompanying ^{252}Cf(s.f.) listing: the first author of the published experiment, location of laboratory, intensity of Cf source, detected radiations after decay of pions or muons, radiation detectors, measured relative branching ratio of neutral pion ($\Gamma_{\pi 0}/\Gamma_{sf}$) or charged pion and muon ($\Gamma_{\pi\pm\ +\ \mu\pm}/\Gamma_{sf}$) emission per ^{252}Cf spontaneous fission and references.

First Author	Year Lab	^{252}Cf f/s	Detected radiations	Radiation Detector	Branching Ratio	Ref.
neutral pion (π^0) radioactivity leading to the high energy gamma production						
J. Beene	1988 ORNL	$5.5\ 10^{+3}$	two γ (> 10 MeV)	63 NaI(Tl) of 17.8 cm thick	$\Gamma_{\pi 0}/\Gamma_{sf}$ $< 1\ 10^{-9}$	[103]
C. Cerruti	1988 Saclay	$1.0\ 10^{+5}$	two γ ($\approx$ 68 MeV)	two 15×15×35cm^3 Pb glasses	$\Gamma_{\pi\pm}/\Gamma_{sf}$ $< 5\ 10^{-9}$	[104]
J. Julien	1989 Modane	$1.0\ 10^{+5}$	two γ (55 –80 MeV)	six 15×15×30cm^3 Pb glasses (undergr.)	$\Gamma_{\pi 0}/\Gamma_{sf}$ $< 1\ 10^{-12}$	[105]
S. Stanislaus	1989 Vancouver	$3.2\ 10^{+6}$	two γ ($\approx$ 68 MeV)	two large NaI	$\Gamma_{\pi 0}/\Gamma_{sf}$ $\approx 3.3\ 10^{-10}$	[106]
J. Knudson	1991 LANL	$6.7\ 10^{+7}$	two γ ($\approx$ 68 MeV)	six Pb-glass + multiwire + calorim.	$\Gamma_{\pi 0}/\Gamma_{sf}$ $< 1.37\ 10^{-11}$	[107]
charged pions (π^+, π^-) and/or muons (μ^+, μ^-) radioactivity						
D. Bucarescu	1987 Bucharest	$9.0\ 10^{+2}$	$\pi^+ \to \mu^+$ $\pi^- \to \mu^-$	six emulsion plates	$\Gamma_{\pi\pm\ +\ \mu\pm}/\Gamma_{sf}$ $< 1\ 10^{-8}$	[108], [109]
Y. Adamchuk	1989 Dubna	$8.3\ 10^{+2}$	$\pi^\pm$, $\mu^\pm \to e^\pm$ (20 –50 MeV)	4 mm thick Plastic + 12 ×15 cm NaI(Tl)	$\Gamma_{\pi\pm\ +\ \mu\pm}/\Gamma_{sf}$ $\approx 5\ 10^{-8}$	[110]
H. Otsu	1992 Tokyo	$6.9\ 10^{+5}$	$\pi^- \to np$ ($\approx$ 70 MeV)	Si solid detector followed by NaI(Tl)	$\Gamma_{\pi -}/\Gamma_{sf}$ < $1.3\ 10^{-8}$	[111]

The bottom part of Table 8.1 additionally lists the few known measurements of the charged pions (π^+ or π^-) or muons (μ^+ or μ^-) emission probability relative to ^{252}Cf spontaneous fission [108] - [111]. In these experiments the primary muons from ^{252}Cf(s.f.) and those produced as a result of charged pions decay are not usually distinguished. It means that measured probability is a sum of the charged pion and muon radioactivities. The most reliable measured upper limits for $\Gamma_{(\mu\pm\ +\ \pi\pm)}/\Gamma_{sf}$ are equal or less than (1 – 5) 10^{-8}. This value is by (3 – 4) orders of magnitude larger than the probability of neutral pion emission in accordance with the energetic consideration and rest masses of leptons. As it was already stated above, the main decay modes of the charged pions and muons do not directly lead to electromagnetic radiation. The 0.511 MeV gammas produced by annihilating positrons, e.g. from decay $\mu^+ \to e^+\ \bar{\upsilon}_e\ \nu_\mu$, will be obviously too negligible contribution to the gamma spectrum from spontaneous fission of ^{252}Cf.

Fig. 8.1 displays the known measured prompt or total γ-ray energy spectra from ^{252}Cf(s.f.) in the energy interval 10 – 200 MeV (most of the data shown were discussed in Section 4.2 and already shown in Fig. 4.3). What has now been added to the plot are the γ-ray spectra from decay of neutral pion emitted by spontaneous fissioning ^{252}Cf with two branching ratios $\Gamma_{\pi 0}/\Gamma_{sf}$ equals 1.37×10^{-11} and 1×10^{-12}, i.e., the minimal upper limits which were measured by J. Julian et al. [105] and J. Knudson et al. [107], respectively. We assumed a Gaussian energy distribution for π^0 decay gammas with mean energy 67.5 MeV and the relative energy spread or resolution FWHM = 30%. The latter roughly reflects the instrumental resolution and the gamma energy range within which the search for the pion decay gammas was carried out in the known experiments. There is no experimental or theoretical information about the energy distribution of emitted π^0 and associated decay gammas.

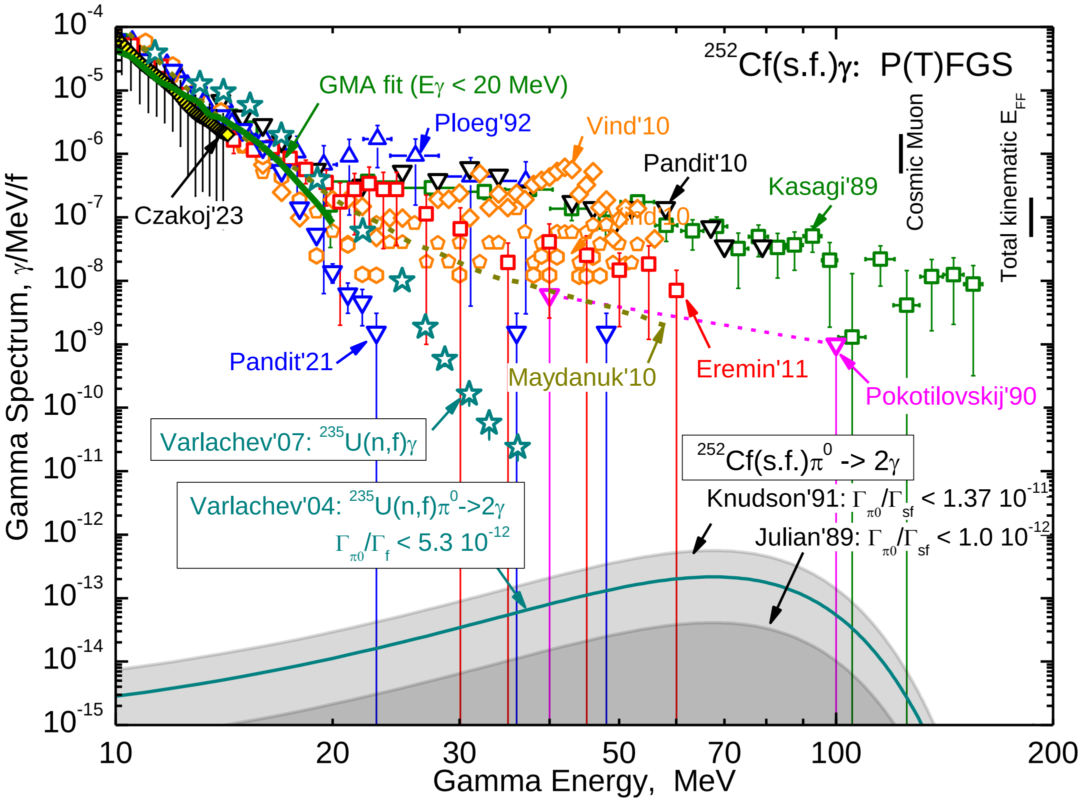

FIG. 8.1. Prompt and total γ-ray energy spectra from ^{252}Cf(s.f.) in the energy interval from 10 to 200 MeV (most experimental and evaluated data are the same as in Fig. 4.3). Additionally, the γ-ray spectrum from decay of neutral pions emitted by spontaneous fissioning of ^{252}Cf are co-plotted as grey curves with $\Gamma_\pi/\Gamma_{sf} = 1.37\times10^{-11}$ [107] and 1×10^{-12} [105]. For comparison, the dark cyan curve shows the γ-ray spectrum for $^{235}U(n_{th},f)\pi^0\rightarrow2\gamma$ with $\Gamma_\pi/\Gamma_f = 5.3\times10^{-12}$ [112].

It is interesting to compare the neutral pion radioactivity followed by decay $\pi^0 \rightarrow 2\gamma$ in the case of ^{252}Cf spontaneous fission with the same data for neutron induced fission. Fig. 8.1 depicts the gamma spectrum computed similarly, but using an upper limit $\Gamma_{\pi0}/\Gamma_f \leq 5.3\ 10^{-12}$ measured for $^{235}U(n_{th},f)$ by V. Varlachev et al. [112], [113]. It can be seen that the absolute emission rate per fission event of neutral pions and, subsequently, of the associated π^0 decay gamma spectra from the ^{252}Cf(s,f.) and ^{235}U(n,f) nuclei are comparable regarding the accuracy of the measured and reported information so far.

More generally, it can be observed in Fig. 8.1 that the prompt or total gamma spectra above 15 MeV measured in the recent underground experiment of D. Pandit et al. for ^{252}Cf(s.f.) γ [46] and at the opened reactor core experiment of V. Varlachev for $^{235}U(n_{th},f)\gamma$ [88] have a similar, exponentially decreasing trend up to (25 – 35) MeV. The similarity likely extends to higher energies, up to (100 – 150) MeV, where both gamma spectra from "classical" fission look transforming in the γ-ray spectra from the decay of neutral pions emitted during fission.

9. Summary

The non-model GLS evaluation of the prompt γ-ray spectrum (PFGS) for spontaneous fission of ^{252}Cf has been performed using the GMA code. This approach provided PFGS in the energy range from 0.1 to 20 MeV with an uncertainty varying between ≈ 2.6 – 22%. However, the obtained reduced chi-square parameter 2.8 points to the possible increasing of the evaluated spectrum uncertainties by factor 1.7.

Above 20 MeV the amplitude and energy shape of the γ-ray spectrum could not yet be established, neither experimentally nor theoretically. The evaluated prompt energy spectrum could be considered as reference. It may complement the prompt fission neutron spectrum for ^{252}Cf(s.f.) which is known with comparable uncertainty and has already been included in the neutron cross section Standards. The precise knowledge of both the neutron and gamma emission spectra is very valuable for diverse fundamental and practical applications with Cf source.

To perform a non-model evaluation of the prompt gamma fission spectrum relevant measurement results were searched in the literature, EXFOR and other databases. 19 experiments were found where the PFGS was measured in coincidence with fission events, 5 experiments where spectra were additionally attributed to the specific fission fragment masses, and 5 experiments where the total gamma spectrum (TFGS) was measured. The latter should be identical to the PFGS above γ-ray energy ≈ 1.8 MeV, i.e., the maximum energy experimentally observed for delayed gamma emission. All these measurements were analysed from the view of suitability for use in the present evaluation (reliable energy range, applied corrections, uncertainties etc.). Additionally, the experimental data on the γ-ray emission multiplicity and average energy were collected and used in the present analyses. 8 experiments were found additionally to those where these integrated quantities could be calculated from the measured energy γ-spectra.

Measurements of the prompt emission of γ- and X-rays with energies lower than 100 keV were found in 4 experiments. Regrettably but they reported the yields of the most intensive (not all) discrete radiation transitions and in the restricted energy range (narrow than 100 keV). This did not allow us to extend the reference PFGS to the energy domain below 100 keV.

A similar deficient status was established for the delayed fission γ-ray spectra (DFGS) for ^{252}Cf(s.f.): we only managed to find 4 relevant experiments. They also do not provide energy differential or integrated delay radiation for the entire energy and time emission ranges. Their results differ substantially from each other, so that they cannot be used as a reliable basis for quantitative evaluation. For the time being, the old evaluation by D. Stoddard could be used in the applications since it partially supported by these measurements and no other alternative exists to date.

From the survey of the existing experimental data it follows that additional precise and complete measurements of the prompt (below 100 keV and above 20 MeV) and delayed (< 1.8 MeV) energy distributions as well as their multiplicities are necessary. This will eventually lead to the complete and precise characterization of all photon emission modes for ^{252}Cf(s.f.).

The analyses of the status of the γ-ray emission data for ^{252}Cf in the major evaluated nuclear data libraries has established the following: ENDF/B-VIII.0 does not have any γ-ray energy spectra from spontaneous fission of ^{252}Cf; JEFF-3.3 adopts the data measured in one single experiment; ABBN-93 only included the total γ-ray spectrum. Not one library contains the delayed γ-ray data and this remains unchanged since 1965 when the evaluation by Stoddard was issued, but without full details and in the too wide energy bins. The major libraries do have information about the X- and γ-ray discrete transitions which follow the α-decay of ^{252}Cf.

The overview of the neutral pionic radioactivity from ^{252}Cf(s,f) indicates that the upper limit for its probability relative to fission is experimentally established at a level (10^{-11} - 10^{-12}). The absolute energy spectrum of the associated γ-rays, which follow the π^0 decay, may be a lowest limit for TFGS in the energy range ≈ (50 - 100) MeV. The same absolute value and energy shape for the high energy gammas were observed for the thermal neutron induced fission of ^{235}U.

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