Investigation of high energy $\gamma$-rays accompanying spontaneous fission of $^{252}$Cf in double and triple neutron-$\gamma$ coincidences

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Abstract. The high energy bremsstrahlung $\gamma$-rays accompanying the spontaneous fission of $^{252}$Cf were measured in the 10-70 MeV energy range. The photons were detected by two BGO scintillator detectors ($\odot 7.6 \text{cm} \times 7.6 \text{cm}$) in coincidence with neutrons detected by plastic scintillator detector, in the 90$^\circ$ and 180$^\circ$ geometry of the two BGO detectors with respect to the axis of the plastic scintillator. The distance from the $^{252}$Cf source to the BGO detectors was 10 cm, and the one to the plastic detector was 50 cm. The fast digital shape analysis technique and the time-of-flight method were used to reject pile-up effects and cosmic ray background. The $\gamma$-ray emission probability of $3 \times 10^{-8} \text{photon/(MeV}\times\text{fission})$ at $E_\gamma = 70 \text{ MeV}$ was obtained.

1. Introduction

One of the intriguing questions in physics of spontaneous fission of heavy nuclei remains the possibility of emission of high energy $\gamma$-rays ($E_\gamma > 20 \text{ MeV}$) accompanying spontaneous fission. The coherent nucleus-nucleus bremsstrahlung mechanism may be responsible for this phenomenon. A similar process of bremsstrahlung emission was observed for the $\alpha$-decay of heavy nuclei. The photon bremsstrahlung spectra for the $\alpha$-decay of the $^{226}$Ra and $^{214}$Po nuclei were measured up to $E_\gamma$ energy of about 1 MeV[1, 2]. In the case of spontaneous fission of heavy nuclei one can expect the emission of high energy $\gamma$-rays because of the bigger Q-value ($Q \sim 200 \text{ MeV}$). The preliminary results of the high energy bremsstrahlung photon emission probability accompanying the binary spontaneous fission of $^{252}$Cf were presented in Ref. [3]. The calculation by the quantum mechanical model presented in Ref. [4] describe in good way the experimental bremsstrahlung spectrum obtained in [3] for binary spontaneous fission of $^{252}$Cf. Moreover, in Fig. 3 (b) of Ref.[4] were also presented the theoretical results of the bremsstrahlung spectrum for the $\alpha$-decay of the $^{226}$Ra, $^{214}$Po, and $^{252}$Cf nuclei, in comparison with the available experimental data of $^{226}$Ra and $^{214}$Po.
The first experimental investigation of high energy $\gamma$-rays was made by the group of Kasagi[5]. They selected the $\gamma$-$\gamma$-coincidences among the emitted $\gamma$-rays accompanying the fission fragments. The observed spectrum had a high energy "tail" which began from $E_\gamma$ about 20 MeV up to about 160 MeV. Authors connected these $\gamma$-rays with the coherent nucleus-nucleus bremsstrahlung emission, affirming instead that at $E_\gamma$ lower than 20 MeV the cascade of $\gamma$-rays and the contribution of $\gamma$-rays due to the giant dipole resonances of excited fission fragments were dominant.

The Kasagi report was so unexpected that many attempts have been made in order to repeat this result. There were two approaches in experimental investigations: i) to measure the exclusive $\gamma$-ray spectrum by means of big volume scintillators with plastic anticoincidence shielding[6, 7]; ii) to measure the high energy $\gamma$-rays by using the $\gamma$-$\gamma$[5, 7] or $\gamma$-fission fragments[7, 8] coincidence techniques. In the energy range $E_\gamma < 20$ MeV there is a reliable agreement between all authors, but above 20 MeV the results are contradictory.

Yu.N. Pokotilovskii[6] measured high energy $\gamma$-rays by using a big volume NaI(Tl) detector with an anticoincidence plastic scintillator covering. The upper limits of the $\gamma$-ray emission probability in the $E_\gamma = 20-140$ MeV energy region of about $6 \times 10^{-9} \text{ photon}/(\text{fission} \times \text{MeV})$ at $E_\gamma = 40$ MeV and $1 \times 10^{-9} \text{ photon}/(\text{fission} \times \text{MeV})$ at $E_\gamma = 100$ MeV were obtained. The $\gamma$-$\gamma$-coincidence method was used by the group of Kasagi[5], and later by the group of Pandit[9]. Their experiments got the same results, in the $E_\gamma = 8-80$ MeV energy range. The relative probability of $\gamma$-ray emission at 20 MeV was about $7 \times 10^{-6} \text{ photon}/(\text{fission} \times \text{MeV})$ and at 80 MeV was about $3 \times 10^{-7} \text{ photon}/(\text{fission} \times \text{MeV})$.

S.J. Luke et al.[7] used the $\gamma$-$\gamma$ and $\gamma$-fission fragment coincidence methods to measure high energy $\gamma$-rays emitted from $^{252}$Cf. The high energy $\gamma$-rays were detected by the NaI(Tl) scintillator with an active anticoincidence shielding. By the first method the integrated yield was $(2.4 \pm 1.3) \times 10^{-6} \text{ photon/fission}$ and by the second one the upper limit of about $1.8 \times 10^{-6} \text{ photon/fission}$ was obtained.

H. van der Ploeg et al.[8] used the $\gamma$-fission fragment coincidence technique. The $\gamma$-ray spectrum was measured by big volume BaF$_2$ and NaI(Tl) detectors. The NaI(Tl) detector was covered by a plastic anticoincidence shielding. By the NaI detector, the determinations of upper limits were $5.7 \times 10^{-7}$ and $2.0 \times 10^{-8} \text{ photon}/(\text{fission} \times \text{MeV})$ in the 20-30 and 30-40 MeV regions, respectively.

In such experiments, the difficulties related to the identification and selection of events included the influence of cosmic ray background events, pile-up effects of gamma-neutron emission multiplicity events and other kind of events on the rate of the true interested events. With the aim to reduce the cosmic ray background we propose a new experimental method based on the neutron-$\gamma$ ($n$-$\gamma$) coincidence. We strongly rejected the pile-up effects by using the digital storage oscilloscope technique and digital processing of signals.

2. Experiment
2.1. Experimental setup
The experimental setup consisted of two BGO scintillator detectors and one plastic scintillator detector connected with a digital storage oscilloscope Tektronix-TDS 7704B. The BGO cylindrical scintillator crystals ($\Theta$76mm $\times$ 76mm) and plastic scintillator ($\Theta$60mm $\times$ 20mm) were coupled with PMTs XP 4312(Photonis Co. Prod.). BGO detectors were used to measure $\gamma$-ray energy. The distance between the $^{252}$Cf source and each of the two BGO detectors was chosen 10 cm in order to avoid multiple taking of $\gamma$ emitted from source. The plastic scintillator detector was able to detect $\gamma$-rays as well as neutrons. The distance between the source and the plastic detector was chosen 50 cm with the aim to distinguish neutrons from photons by the time-of-flight (TOF) method. The axes of the two BGO detectors in respect to the axis of the plastic detector formed the angles 90° and 180°, respectively.
The software code developed for the Tektronix-TDS 7704B allowed us to detect double and triple coincidences within the 200 ns time window between the signal from plastic detector and one (in double coincidence mode) or two signals (in triple coincidence mode) from the BGO detectors, and to save the shapes of pulses from all detectors in the oscilloscope memory. We used signals from plastic detector as ”start” and the signals from BGO detectors as ”stop”. Experimental data were collected in a continuous mode during the total interval of 78 days of data taking. The off-line processing of signals was made by a personal computer. The activity of the $^{252}$Cf source was $6.1 \times 10^6$ neutron/s.

The absolute efficiency of the $\gamma$-ray detecting system was calculated by GEANT4. In simulation, the source of $\gamma$-rays was modelled in the geometry of our experimental setup. We assume the absolute efficiency of detector as the number of photons which lost almost all its energy (with accuracy of the energy averaging interval) in the BGO detector per the total number of $\gamma$-rays emitted from the source. For $\gamma$-rays of 70 MeV leaving 65-70 MeV of its energy the absolute detecting efficiency of BGO was about 0.2%.

2.2. Data processing
The goal of data processing was to determine the energy of pulses in both BGO detectors and to estimate the delay time between the pulses of the plastic detector and the BGO detectors in the coincidence modes. We used the delay time to distinguish the n-$\gamma$ coincidences from the $\gamma$-$\gamma$ ones and the cosmic shower coincidences.

The data processing included the rejection of pile-up events to be sure that we measured the $\gamma$-ray energy accurately. The registered pulses (voltage versus time) were smoothed and local extremums were found for each pulse. If in neighbourhood of such local extremum the pulse area at the right side of the considered extremum was greater than the one at the left side, then this pulse was considered as a superposition of pulses and rejected.

To calculate the delay time we found the time marker position for each pulse. The marker position was found by the constant fraction discriminator method at $1/3$ of the pulse amplitude. The amplitudes of pulses were found for the smoothed shapes of signals. To obtain the energy of BGO pulses, we calculated the pulse area by using the coefficients of the calibration of the BGO detectors.

2.3. Calibration
To calibrate each of the BGO detectors we used standard radioactive sources as $^{137}$Cs, $^{60}$Co and Pu(Be), and cosmic muons. The experimental setup for the calibration by muons is presented in Fig.1 (left side), and the BGO detector was in coincidence with the plastic detector. We calculated by GEANT4 the energy lost by cosmic muons in the BGO scintillator, and the results of calculation (triangles) in comparison with experimental data (circles) are presented in Fig.1 (right side). The experimental and theoretical spectra have the same maximum near 70 MeV.

3. Results and discussion
3.1. Time-of-flight spectrum
The main idea of our experiment was the use of the TOF method to reduce the influence of cosmic background by means of the delay time between the pulses in the plastic detector and the BGO detectors, so that we could choose the n-$\gamma$ coincidences from the ones which also comprised the cosmic shower coincidences. The typical shape of the TOF spectrum is presented in Fig.2. The time resolution of the $\gamma$-$\gamma$ peak was about 2 ns. Cosmic rays gave a contribution in two ways. Near the peak of $\gamma$-$\gamma$ coincidences there is a peak of cosmic shower coincidences which shows a Gaussian-like distribution. The shift between the top of $\gamma$-$\gamma$ peak and the top of cosmic peak is about 2 ns. This is needed time to a $\gamma$-photon to pass 50 cm from the $^{252}$Cf source to
the plastic detector. Also there were random coincidences between cosmic rays and neutrons emitted from the source, and between cosmic rays and $\gamma$-rays emitted from the source. These events show an uniform distribution.

To estimate the influence of cosmic ray events we made measurements with the same geometry but without source. The total rate of high energy $\gamma$-rays ($E_\gamma > 30$ MeV) was $1.7 \times 10^{-5}$ photon/(MeV $\times$ s) in the region of $\gamma$-$\gamma$ coincidence peak. In the experiment with the $^{252}$Cf source the rate of $\gamma$-rays ($E_\gamma > 30$ MeV) from n-$\gamma$ coincidences was $1.5 \times 10^{-6}$ photon/(MeV $\times$ s). But the peak of n-$\gamma$ coincidences cross only the tail of the peak of cosmic coincidences, so cosmic events may give a contribution lower than about 1%. The final estimation of the rate of cosmic coincidences was lower than $1.7 \times 10^{-7}$ photon/(MeV $\times$ s) in the region of n-$\gamma$ peak.

3.2. Emission probability and result

In the TOF spectrum we selected one time window in the region of n-$\gamma$ coincidences and another one of the same time duration is symmetrically chosen at the left side with respect to the peak of cosmic shower coincidences. In the second region there are only random coincidences. In order to subtract the contribution of cosmic ray background from the n-$\gamma$ coincidences, we subtracted the spectrum of the random coincidences (left window) from the true n-$\gamma$ coincidences plus the random spectrum (right window). The probability of $\gamma$-ray emission accompanying spontaneous fission of $^{252}$Cf was calculated by formula:

$$P_\gamma(E_\gamma) = \frac{N_{n-\gamma}(E_\gamma)}{\epsilon_{\gamma}(E_\gamma) \cdot T \cdot N_n \cdot \Delta E_\gamma}$$

(1)

where $P_\gamma(E_\gamma)$ is the probability of photon emission with energy $E_\gamma$; $N_{n-\gamma}(E_\gamma)$ is the number of the registered n-$\gamma$ coincidence events with photons of energy $E_\gamma$; $\epsilon_{\gamma}(E_\gamma)$ is the absolute efficiency of the BGO detector which registers photons with energy $E_\gamma$, calculated by GEANT4; $T$ is the total time of the collected coincidence events; $N_n$ is the number of registered pulses by the plastic detector in single mode; $\Delta E_\gamma$ is the interval of energy for the averaging procedure.

The obtained probability is presented in Fig.3. For 90° and 180° geometry of BGO, the
Figure 2. TOF spectrum of $^{252}\text{Cf}$. The labels denote: 1 - the peak of cosmic showers; 2 - the peak of $\gamma-\gamma$ coincidences; 3 - the peak of the n-$\gamma$ coincidences; 4 - the random coincidences. Dashed line represents the approximation of peaks 1 and 2 by sum of two Gaussian curves. The Gaussian representing the peak of the cosmic showers is denoted by full line.

Figure 3. Photon emission probability of the high energy $\gamma$-rays accompanying spontaneous fission of the $^{252}\text{Cf}$ nucleus.

emission probability values at high energy $\gamma$-rays were of the same order. In comparison with the results of Kasagi[5] and Pandit[9] groups, our obtained probability values are about one order lower at $E_\gamma \sim 30$ MeV and become equal at $E_\gamma \sim 70$ MeV. The relevant difference between the
obtained probabilities at $E_\gamma \sim 30$ MeV of other groups and our presented results may be due to their incomplete cut off of the cosmic ray background leading to an overestimation of the $\gamma$-emission probability by the $\gamma-\gamma$ coincidence method.

Pokotilovskii\cite{6} and Luke\cite{7} used complicated active and passive shieldings in order to reduce cosmic ray background. For our opinion, they may reduce also true coincidence events in the case of high energy $\gamma$-rays, because such $\gamma$-rays do not lose all its energy in the $\gamma$-detector since they lost some part in the anticoincidence shielding. In Fig.3 our results reproduce the estimated upper limits for the high energy $\gamma$-rays presented in Refs. \cite{6, 7}. Therefore, only at $E_\gamma \sim 70$ MeV our results are in good agreement with the determinations of Kasagi and Pandit, and with the upper limit given by Luke.

3.3. Triple coincidences

Fig. 4 shows our preliminary results of triple coincidences between pulses of the two BGO detectors and plastic detector. The peak of the $\gamma-\gamma-\gamma$ coincidences and the distribution of the $n-\gamma-\gamma$ coincidences are clearly resolved. The $n-\gamma-\gamma$ coincidence distribution presents a peak (at the typical delay time of about 20 ns) corresponding to neutrons with energy of about 5 MeV. Also a local maximum (indicated in figure by the upper arrow) along the $n-\gamma-\gamma$ coincidence distribution, is observed at the delay time of about 10 ns, corresponds to neutrons with energy of about 10 MeV. In the right side of figure are reported only the events of triple coincidences with $E_\gamma > 10$ MeV only.

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