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Fragment dependence of high energy γ-ray emission in the spontaneous fission of ^{252}Cf

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Abstract. The high energy γ-ray emission accompanying the spontaneous fission of ^{252}Cf has been measured in coincidence with individual fission fragments selected by discrete γ-ray transitions. The enhancement of the γ-ray emission probability in the energy range E_γ=3-8 MeV has been observed for the fission fragments in the region of nearly symmetric mass splitting, confirming results reported in previous investigations. The γ-γ coincidence technique employed in the present work clearly demonstrate that the major contribution to this enhancement is caused by the fission channels where one fragment is near to the N=82 or Z=50 shell closures. The high energy γ-ray emission probability does not show any significant dependence on the number of neutrons emitted in the fission process, supporting the hypothesis that high energy γ-rays are mainly emitted from the fragments after the neutron evaporation.

PACS. 25.85.Ca Spontaneous fission – 23.20.Lv Gamma transitions and level energies – 27.60.+j $90 \leq A \leq 149$

1 Introduction

There has been a renewed interest in the study of high energy γ-ray emission accompanying the spontaneous fission of ^{252}Cf after the observation of a pronounced enhancement in the γ-ray energy spectrum at E_γ=3-8 MeV [1]. Since then, several measurements of ^{252}Cf fission have been carried out to investigate the dependence of the high energy γ-ray emission on the fragment mass in binary [2–4] as well as in α-particle accompanied fission [5]. The results of those studies show that the enhancement in the γ-ray energy spectrum is mainly correlated with near mass symmetric (132:120) fission channels. Furthermore, this enhancement was found to be associated predominantly with the heavier partner and to have an almost isotropic angular distribution.

The experimental trends were reproduced [2] by statistical model calculations performed with the CASCADE code [6], in which an experimental mass dependent level density parameter a, obtained from the study [7] of the neutron emission from the same ^{252}Cf source, was used. This level density parameterization shows effects related to the shell corrections which are maximized in nuclei at the shell closures. An important point, suggested by the statistical model analysis, is that the enhancement in the γ-ray emission is correlated essentially to nuclear level density effects, being not sensitive to the excitation energy of the initial fission fragments at scission.

Although important progress have been obtained in the understanding of the high energy γ-ray emission in the spontaneous fission of ^{252}Cf, some questions have still to be experimentally addressed. The experiments performed so far employed kinematical coincidence methods using gas detectors. The fragment masses were determined with a resolution of about 4-10 amu [FWHM] and the high energy γ-ray spectra were obtained for mass bins 10 amu wide [2–5]. Therefore, the specific fragments responsible for the effect have not completely identified. Furthermore, it is experimentally known that the average neutron multiplicity is very low [7] for fragments near the shell closures (Z=50 and N=82), where the bulk of the high energy γ-ray enhancement is supposed to take place. This observation raises the question whether the reduction in the number of emitted neutron in the fission process should be correlated with the enhancement of high energy γ-rays. The study of the neutron-gamma correlation would confirm that the high energy γ-rays are predominantly emitted after the neutron emission, as suggested by Statistical Model calculations.

A new investigation of the high energy γ-ray emission accompanying the spontaneous fission of ^{252}Cf has been now performed in which the fission fragments have been selected by gating on their known low energy γ-ray transitions by using the GASP spectrometer. This technique allows a direct tagging of at least one of the two fission fragments and in this way, the atomic numbers as well as the atomic masses of the fragments are determined. It has to be mentioned that γ-γ coincidence technique has been found to be extremely suitable not only to identify the fission fragment and study their spectroscopy but also to determine the number of neutrons emitted in the ^{252}Cf spontaneous fission [8].

In this paper the results of this new study of the spontaneous fission of ^{252}Cf are presented. Experimental details are given in Sect. 2. The data relative to the high energy γ-ray yield in coincidence with a number of fission fragments are presented and discussed in Sect. 3. Conclusions are drawn in Sect. 4.

2 Experimental setup

The experiment was performed at the GASP spectrometer [9] located at the Tandem Laboratory of the Laboratori Nazionali di Legnaro. In its standard configuration, GASP consists of an array of 40 large volume Compton suppressed germanium detectors (HPGe), positioned in 7 rings at different angles with respect to the beam direction, and of an inner ball of 80 bismuth germanate scintillators (IBBGO) having a thickness of 6 cm. In the present experiment, energy and time signals of the individual IBBGO scintillators were recorded in order to detect γ-ray up to $E_\gamma \sim 20$ MeV, as already done in past work [10]. Furthermore, two of the HPGe detectors of the spectrometer (at $\theta = 90°$ and $145°$) have been replaced by large volume (10 cm × 10 cm) cylindrical BGO crystals (LBGO), which were temperature stabilized and gain monitored, in order to obtain high resolution spectra of energetic γ-rays. Both scintillators were positioned at 75 cm from the target. The time-of-flight technique, with the start signal given by the GASP inner ball, was used to distinguish γ-rays from neutrons. A complete description of the operation of the LBGO detectors in coincidence with the GASP spectrometer can be found in [11].

A sealed ^{252}Cf source ($\sim 10^4$ fission$\times$s^{-1}) was placed at the centre of the GASP array. The total counting rate of the inner ball was 15 kHz only. Events were collected during a 3 weeks run with the condition of having at least two inner ball elements in coincidence with one HPGe detector. The LBGO detectors signals were recorded whenever they fired.

The energy calibrations of the detectors were obtained by using standard low energy radioactive sources. In particular, the LBGO and IBBGO detectors were calibrated using ^{88}Y (E_γ=0.898, 1.836 MeV) and ^{56}Co (E_γ=2.598, 3.243 MeV) sources. Moreover, the energy calibrations were performed periodically during the experiment to monitor the stability of the IBBGO detectors due to temperature and/or electronic drifts. As a result, the maximum variation of the channel position corresponding to the E_γ=3.243 MeV peak of ^{56}Co was observed to be ±3% of its average value considering the 80 IBBGO detectors over the complete run.

In the off-line analysis prompt HPGe γ-rays events were selected by requiring a gate of $\Delta t \approx 40$ ns in the TDC spectra started by the inner ball. The HPGe data were sorted to build E_γ-E_γ matrices and E_γ-E_γ-E_γ cubes. E_γ-E_γ-E_γ^{LBGO}(E_γ^{IBBGO}) cubes were also generated where E_γ^{LBGO} (E_γ^{IBBGO}) is the energy measured in individual LBGO (IBBGO) detectors. A total statistics of 0.8 × 10^9 events were obtained having two prompt γ-rays detected in HPGe and one γ-ray with $E_\gamma \geq 1$ MeV in the IBBGO individual detectors. This figure reduces by about two orders of magnitude, when the events in the external LBGO are considered.

It has to be noted that neutron induced events are not suppressed in the IBBGO spectra, being the flight path too short (17 cm) to reject them by the time-of-flight technique. However, the neutron-to-γ ratio has been obtained from the time of flight measurement of LBGO detectors. This information was used to estimate the neutron contamination in the IBBGO spectra which resulted to be quite low, due to the reduced efficiency ($\epsilon_n \sim 10\%$) of such scintillators for neutrons [12]. As a conclusion, the neutron induced events are supposed to weakly influence the measured energy spectra. Furthermore, this contamination is negligible when the energy spectra associated with different fragment mass splits are compared.

The selection of a single fission fragment was performed by gating on low energy γ-ray transitions in the HPGe spectra. The choice of the transitions has been done by verifying in the E_γ-E_γ matrix the absence of contaminations from other nuclei in a given energy gate. However, the large number of nuclei produced and the lack of information on the level scheme of several fission products suggested in general to tag each nucleus by using at least two coincident transitions. This double gating procedure reduces the available statistics, especially in the case of high energy γ-ray events in the external LBGO detectors. Consequently, the latter data were used only to cross check the results from the IBBGO detectors which are presented in the following. It is important to note that the spectra and the relative yield data reported in this work are not corrected for the detector efficiency.

3 Experimental results

As an example of the data obtained in the present experiment, we present in Fig. 1 the high-energy γ-ray spectra in coincidence with ^{142}Ba, ^{144}Ba, ^{136}Xe, ^{135}I and ^{134}Te nuclei. It is observed that the shape of the spectra depends strongly on the selected fission fragment. In some cases, as those of the ^{142}Ba and ^{144}Ba nuclei, the spectral shape exhibits the well known exponential fall-off characteristic of the statistical E1 emission. On the contrary, in the case of ^{136}Xe, ^{135}I and ^{134}Te nuclei a clear enhancement at E_γ=3-8 MeV is observed, with respect to the exponential shape of the ^{144}Ba nucleus. As clearly seen from Fig. 1, the size of the enhancement depends on the selected fragment.

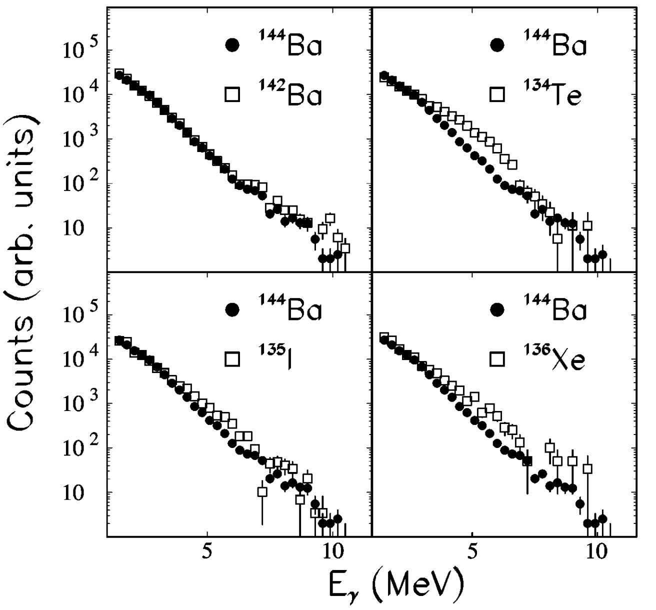

Fig. 1. The γ-ray energy spectra in coincidence with ^{136}Xe, ^{135}I, ^{134}Te ^{142}Ba and ^{144}Ba fission fragments. The spectra have been obtained by double gating the E_γ-E_γ-E_γ^{IBBGO} cube on the γ-ray transitions reported in Table 1

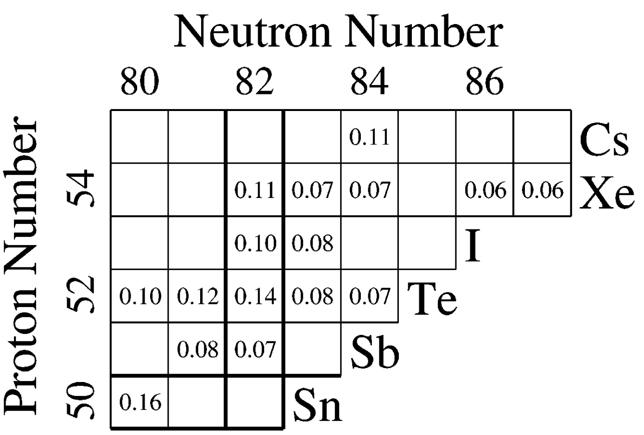

Fig. 2. The ratio R for various heavy fission fragments produced in the spontaneous fission of ^{252}Cf. For the definition of R see Table 1

To study this effect in a quantitative way, the ratio R of a given fission fragment in coincidence with an energetic γ-ray ($E_\gamma^{IBBGO} \geq 3.5$ MeV) to the total coincidence yield ($E_\gamma^{IBBGO} \geq 1$ MeV) was determined. Data are presented in Table I and plotted in the N-Z plane in Fig. 2 for various fission channels.

It is observed from Table 1 that products from asymmetric mass splitting, such as the Barium isotopes, show a ratio $R \sim 6\times10^{-2}$. The R value increases significantly up to $R \sim 1.5 \times10^{-1}$ for some nuclei near the N=82 or Z=50 shell closures, thus confirming experimentally the hypothesis from [3]. It appears clearly from Fig. 2 that the value of the ratio R is higher for nuclei north-west from the double shell closure in ^{132}Sn. The value of the ratio decreases, indeed, as one goes away from the N=82 shell closure towards east in the N-Z plane.

Table 1. The ratio R of heavy fission fragments in coincidence with an energetic γ-ray ($E_\gamma^{IBGO} \geq 3.5$ MeV) to the total coincidence yield ($E_\gamma^{IBGO} \geq 1$ MeV). The R-values are reported for various nuclei observed in spontaneous fission of ^{252}Cf. The γ-ray transitions used to identify the nuclei by double gating the E_γ-E_γ-E_γ^{IBBGO} cube are listed as 1^{st} and 2^{nd} gates. For details see the text

Nucleus	1^{st} gate (keV)	2^{nd} gate (keV)	Ratio R
^{139}Cs	219	491	0.097± 0.005
^{136}Xe	1313	381	0.106± 0.004
^{137}Xe	1220	400	0.072± 0.002
^{138}Xe	589	484	0.066± 0.001
^{140}Xe	377	458	0.064± 0.001
^{141}Xe	370	516	0.061± 0.002
^{135}I	1133	288	0.098± 0.002
^{136}I	1111	261	0.075± 0.002
^{132}Te	974	697	0.091± 0.004
^{133}Te	1096	404	0.118± 0.009
^{134}Te	1279	297	0.142± 0.002
^{135}Te	1180	325	0.082± 0.003
^{136}Te	606	423	0.070± 0.001
^{132}Sb	1773	1025	0.077± 0.010
^{133}Sb	2792	1510	0.070± 0.010
^{130}Sn	1221	774	0.158± 0.010
^{142}Ba	360	475	0.063± 0.001
^{143}Ba	117	343	0.067± 0.002
^{144}Ba	199	331	0.063± 0.001
^{145}Ba	112	350	0.055± 0.010
^{146}Ba	181	333	0.061± 0.002
^{147}Ba	110	250	0.051± 0.010

The selection of one fission fragment (FF1) leaves undefined the complementary one (FF2). From previous experiments [13] it is known that, for a given FF1 fragment, only few complementary FF2 fragments would contribute to the measured ratio R, having masses A_{FF2}=252-A_{FF1}-ν_n which are correlated to the number of evaporated neutrons ν_n=2-5. This fact suggests that the enhancement in the γ-ray energy spectrum, as seen from the data reported in Table 1, might be mainly due to the selected heavy fragment [2].

The enhancement, however, can also be observed by tagging the light complementary fragment, as demonstrated in Fig. 3 where the γ-ray spectra are reported selecting the most probable complementary fragments of ^{144}Ba (^{104}Mo and ^{105}Mo) and ^{134}Te (^{114}Pd and ^{116}Pd). These spectra are again compared to that from the ^{144}Ba nucleus as a reference. The R values measured for the complementary light fragments are listed in Table II. As seen in Fig. 3, the light Pd fragments also show the enhancement as their complementary Te fragments do. Similarly, the enhancement is not observed in case of Mo isotopes, as expected from the data relative to their complementary heavy Ba fragments.

The correlation between complementary fragments is of special interest in the case of Sn nuclei for which only

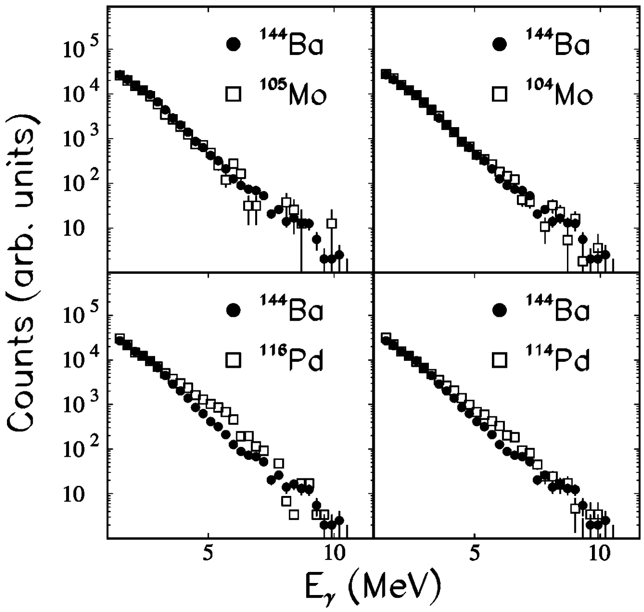

Fig. 3. As Fig. 1 but for light Mo and Pd fission fragments. The spectrum in coincidence with ^{144}Ba is shown as a reference

Table 2. As Table 1 but for light fission fragments

Nucleus	1^{st} gate (keV)	2^{nd} gate (keV)	Ratio R
^{102}Mo	296	584	0.056± 0.002
^{103}Mo	145	331	0.064± 0.002
^{104}Mo	192	369	0.066± 0.001
^{105}Mo	145	419	0.068± 0.001
^{106}Mo	172	351	0.066± 0.001
^{107}Mo	123	348	0.059± 0.005
^{108}Mo	193	371	0.053± 0.002
^{112}Pd	349	535	0.062± 0.001
^{114}Pd	333	520	0.081± 0.001
^{116}Pd	340	538	0.105± 0.001
^{116}Cd	513	706	0.120± 0.004
^{118}Cd	488	677	0.140± 0.003
^{120}Cd	506	697	0.134± 0.004

the ^{130}Sn isotope was directly identified from known γ-ray transitions. In fact, heavier Sn isotopes were not directly detected because their first excited states are located at energies of about 4 MeV, which is beyond the dynamic range of our HPGe detectors. Following the above observation, the enhancement in the γ-ray emission can be checked in this case by looking to the complementary Cd isotopes. In fact, as shown in Fig. 4 and in Table 2, significant enhancement is seen for both ^{118}Cd and ^{120}Cd isotopes.

A second result obtained in the present work is about the relationship between neutron and high energy γ-ray emission in the spontaneous fission. Measured R values relative to the Ba isotopes reported in Table 1 allow a direct test of the proposed insensitivity of the γ-ray emission to the excitation energy of the initial fission fragments at scission. In fact, measured data do not show significant variation when different isotopes ranging from ^{142}Ba to ^{147}Ba are considered. Those isotopes are known to be in coincidence with a substantially different neutron multiplicity (ν_n=1.4-4.3). The reconstructed [8] average excitation energy in Ba isotopes is also supposed to change as a function of the neutron number, being around 10 MeV in ^{144}Ba,^{146}Ba and around 20 MeV in ^{142}Ba,^{147}Ba, respectively. From this point of view it seems that the average excitation energy of the fission fragments (and the correlated neutron multiplicity) does not play any significant role in the high energy γ-ray emission.

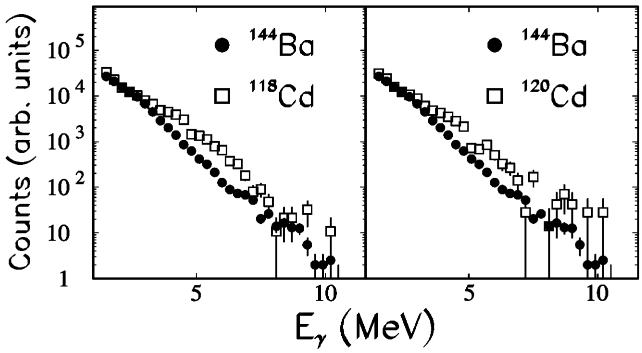

Fig. 4. As Fig. 1 but for light Cd fission fragments, the complementary products of Sn isotopes. The spectrum in coincidence with ^{144}Ba is shown as a reference

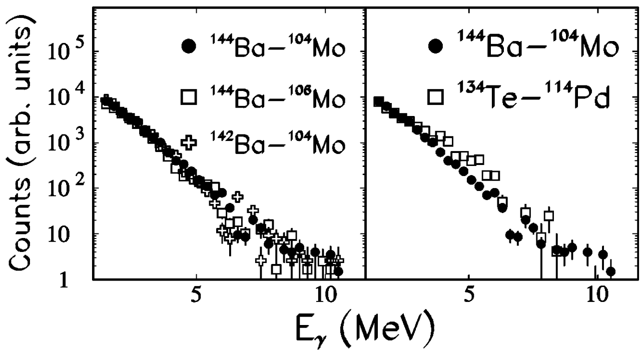

Fig. 5. Left panel: γ-ray energy spectra for Mo-Ba pair in case of 2n, 4n and 6n emission channel; Right panel: comparison of the 4n emission channel in the case of the Mo-Ba and Te-Pd partitions

This problem is illustrated in more detail in Fig. 5, where high energy γ-ray spectra are shown for different selections of the final Mo-Ba fragments which are associated to a different neutron multiplicity. The spectra in Fig. 5 were obtained by carefully selecting two energy gates, one in each fission fragment. It is observed that the high energy γ-ray spectra do not show any significant change going from the 2n (^{144}Ba - ^{106}Mo), to the 4n (^{144}Ba - ^{104}Mo) and to the 6n channels (^{142}Ba -^{104}Mo) emission. On the contrary, the enhancement is clearly seen by comparing asymmetric (^{144}Ba - ^{104}Mo) and nearly symmetric (^{134}Te -^{114}Pd) 4n fission channels.

The results reported here confirm that the high energy γ-rays are emitted predominantly in the later stages of the fragment de-excitation after the cooling due to the neutron evaporation. In fact, it is expected that the emission of energetic γ-ray from the highly excited nuclei would be enhanced by selecting de-excitation chains with a lower number of emitted neutrons [14]. Such correlation is not observed here suggesting that the enhancement can be mainly due to the structure properties of the final nuclei. In particular, as argued in [2], the major role should be played by the level density and its excitation energy dependence, which drives the neutron-gamma competition at excitation energies close to the yrast line.

Statistical Model calculations have been performed, considering as a test case the Sn-Cd splitting, to further test the sensitivity of the γ-ray spectra to the level densities in a given fission channel. Following [2], the ^{131}Sn and ^{121}Cd primary fragments were supposed to be at average excitation energies of $E_{x,1}$=12 MeV and $E_{x,2}$=30 MeV and average angular momenta J_1=J_2=6$\hbar$. The statistical de-excitation of the fission fragments was then modelled using the CASCADE code with an excitation energy independent average level density parameter a=A/8 MeV^{-1} as well as the Reisdorf parameterization for the level density [15]. The latter parameterization takes into account explicitly the shell corrections and its excitation energy dependence in the nuclei of the whole de-excitation cascade. We note that in the original CASCADE code the option is included of a change in the level density parameterization from the "high energy" Fermi gas value for the parameter a to the "low energy" one, where the Dilg results [16] are used. However, this modelling of the level density produces discontinuities which may lead to unphysical results [17].

Calculations including the two prescriptions of the level densities predict as final fragments ^{130}Sn and ^{118}Cd after the evaporation of 1n and 3n, respectively. The calculated γ-ray spectra show that when the average level density parameter a=A/8 MeV^{-1} is used, the emission of γ-rays with energy around E_γ~5 MeV is dominated by the light Cd fragment, which is at higher excitation energy. When the Reisdorf parameterization is used, the Cd spectrum does not exhibit significant variation with respect to the a=A/8 MeV^{-1} case, being the shell corrections small for those nuclei. On the contrary, the Sn spectrum features a sizeable enhancement around E_γ~5 MeV correlated to the large shell corrections for Sn nuclei calculated within the Reisdorf parameterization.

The Statistical Model calculations discussed here are in qualitative agreement with our experimental results showing the importance of the shell corrections to the level density in explaining the γ-ray enhancement.

4 Conclusions

The results reported in this work show that the enhancement in the emission of high-energy γ-rays in spontaneous fission of ^{252}Cf depends strongly on the the mass splitting as already reported in earlier works employing a different experimental technique [1–5]. As a further step in the understanding of these phenomena, it is shown here experimentally that this enhancement is maximized when one of the two fission fragment is near the shell closures at Z=50 and N=82. The small variation of the relative probability of the high energy γ-ray emission in the case of Ba isotopes confirms its suggested insensitivity to the excitation energy of the final fission fragment. Furthermore, the present results do not show a significant dependence of the high energy γ-rays emission probability on the number of neutron emitted in the fission process for the Mo-Ba partition. This implies that in the ^{252}Cf spontaneous fission the excited fragments are mainly cooled down by neutron evaporation, being the high energy γ-rays produced in the latter stage of fragment de-excitation.

Because of the limited quality of the high energy γ-ray spectra measured with the GASP inner ball detectors and the uncertainty on the untagged complementary fission fragment, the present data does not allow the study of this effect in a full quantitative way. Nevertheless, it is clear that the enhancement in the emission of energetic γ-rays is related to the level density parameter a. In fact, as discussed in details in a previous work [2], the level density of closed shell nuclei will present a lower number of levels for bin of excitation energy, thus increasing the probability of energetic γ-ray emission. Statistical Model calculations with the Reisdorf parameterization for the level density nicely illustrate this effect in the case of the Sn-Cd splitting.

As a result, the measure of high energy γ-ray spectra in coincidence with selected fission fragments will open the possibility of a direct test of the level density parameterization of the nuclei at low excitation energies, which is of great importance also for its astrophysical implications [18]. Furthermore, a detailed knowledge of the level density parameter a in nuclei around the N=82 and Z=50 nuclei, will represent an important test for the theoretical description of the nuclear shell model properties [19].

Future prospects of this investigation would require upgraded capability of both low-energy and high-energy γ-ray detection systems. In particular, the use of more powerful arrays, as Euroball and Gammasphere, will allow, as demonstrated recently, the tagging of both fission fragments [13]. This will offer the possibility to perform a more direct comparison with model predictions.

References

1. P. Glassel, R. Schmid-Fabian, D. Schwalm, D. Habs and H. U. v. Helmolt, Nucl. Phys. A502, 315c (1989)
2. H. van der Ploeg, J. C. S. Bacelar, A. Buda, C. R. Laurens, A. van der Woude, J. J. Gaardhoje, Z. Zelazny, G. van 't Hof and N. Kalantar- Nayestanaki, Phys. Rev. C52, 1915 (1996)
3. J. B. Fitzgerald, D. Habs, F. Heller, P. Reiter, D. Schwalm, P. Thirolf, A. Wiswesser, Z. Phys. A355, 401 (1996)
4. A. Hotzel, P. Thirolf, Ch. Ender, D. Schwalm, M. Mutterer, P. Singer, M. Klemens, J. P. Theobald, M. Hesse, F. Gonnenwein, H. v. d. Ploeg, Z. Phys. A356, 299 (1996)
5. P. Singer, M. Mutterer, Yu. N. Kopach, M. Klemens, A. Hotzel, D. Schwalm, P. Thirolf, M. Hesse, Z. Phys. A359, 41 (1997)

6. F. Puhlhofer, Nucl. Phys. A280, 267 (1977)
7. C. Butz-Jorgensen and H. H. Knitter, Nucl. Phys. A490, 307 (1988)
8. G.M. Ter-Akopian et al., Phys. Rev. C55, 1146 (1997)
9. D. Bazzacco et al., Phys. Lett. B309, 235 (1993)
10. L.H. Zhu et al., Phys Rev C55, 1169 (1997)
11. G. Viesti et al., Nucl. Phys. A604, 81 (1996)
12. G. Hausser, M.A. Lone, T.K. Alexander, S.A. Kushneriuk and J. Gascon, Nucl. Instr. Meth. 213, 301 (1983)
13. J. H. Hamilton et al., Prog. Part. Nucl. Phys., Vol. 38, 273 (1997) and references therein
14. A. Atac et al., Phys. Lett. B252, 545 (1990)
15. W. Reisdorf, Z. Phys. A300, 227 (1981)
16. W. Dilg, W. Shantl, H. Vonach and M. Uhl, Nucl. Phys. A217, 269 (1973)
17. J.P. Lestone, Phys. Rev. C53, 2014 (1996)
18. P. Donati, P.M. Pizzocchero, P.F. Bortignon and R.A. Broglia, Phys. Rev. Lett. 72, 2835 (1994) and references therein
19. see e.g.: P. Bhattacharyya et al., Phys. Rev. C56, R2363 (1997) and C.T. Zhang et al., Phys. Rev. Lett. 77, 3743 (1996)