Measurements of prompt fission gamma-rays and neutrons with lanthanide halide scintillation detectors

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Abstract
Photons have been measured with lanthanide halide scintillation detectors in coincidence with fission fragments. Using the time-of-flight information, reactions from γ-rays and neutrons could easily be distinguished. In several experiments on ^{252}Cf(sf), ^{235}U(n$_{th}$,f) and ^{241}Pu(n$_{th}$,f) prompt fission γ-ray spectra characteristics were determined with high precision and the results are presented here. Moreover, a measured prompt fission neutron spectrum for ^{235}U(n$_{th}$,f) is shown in order to demonstrate a new detection technique.

1 Introduction

A good knowledge of particle emission in fission is essential for the peaceful use of nuclear power. Prompt gamma-rays contribute considerably to the fission heat in a reaction core, whereas prompt neutrons are responsible for maintaining a chain reaction. The precision, with which their characteristics are known, is of course important for both safety reasons and economy. Apart from the technological aspects, there are also indications that in particular prompt fission gamma-rays reveal detailed information on the dynamics of the fission process.

A coordinated research program for the experimental investigation of prompt fission neutron spectra (PFNS) from major actinides, induced by fast neutrons, was launched by the IAEA [1], while new measurements of prompt fission γ-ray spectra (PFGS), in particular for the reactions ^{235}U(n,f) and ^{239}Pu(n,f), have been included in OECD-NEA's high priority request list for prompt fission γ-ray data [2]. In recent studies we developed a technique, which allows in principle to measure simultaneously both prompt neutrons and γ-rays emitted in fission, and applied it to the reactions ^{252}Cf(sf), ^{235}U(n$_{th}$,f) and ^{241}Pu(n$_{th}$,f). Below we report on the experiments, recently performed in the framework of ERINDA, and present results that were obtained so far.

2 Experiments

All experiments described in this paper have in common that γ-rays were measured in coincidence with fission fragments. The photons were detected with novel lanthanide halide scintillation detectors, based on cerium-doped lanthanum-chloride ($LaCl_3$:Ce) [3], cerium-doped lanthanum-bromide ($LaBr_3$:Ce) [4] and cerium-bromide ($CeBr_3$) [5, 6] crystals, respectively, which combine an excellent timing resolution with a reasonably good energy resolution. The trigger was provided by the fission fragments, which were detected by either an ultra-fast polycrystalline chemical vapor deposited (pcCVD) diamond detector [7] in conjunction with the fission fragment spectrometer VERDI [8] or a Frisch-grid ionization chamber. In all experiments both energy and time-of-flight (TOF) of the photons were recorded. Figure 1 shows a typical two-dimensional presentation of photons measured with a 2 in. $\times$ 2 in. $LaBr_3$:Ce detector in

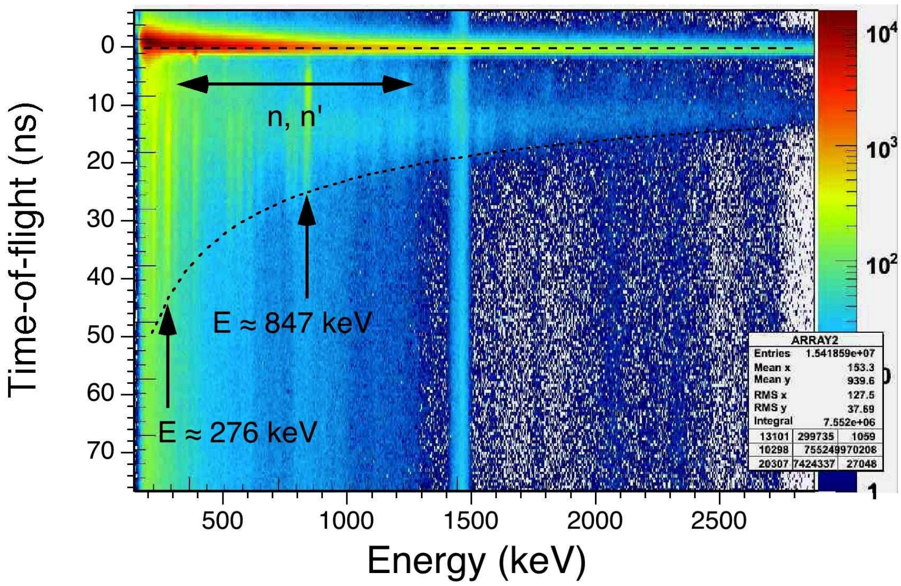

Fig. 1: Distribution of γ-rays by time-of-flight vs. energy. Prompt fission γ-rays are represented by a horizontal area (dashed line), while the vertical lines represent γ-rays from reactions induced by prompt fission neutrons, preferably inelastic scattering (see text for details).

coincidence with fission fragments, here from the spontaneous fission of ^{252}Cf, investigated at the DG Research Centre Institute for Reference Materials and Measurements (IRMM). The region of prompt fission γ-rays is indicated by the dashed line, while the many vertical lines between the dashed and the dotted lines correspond to de-excitations after inelastic neutron scattering in the detector or in structural materials in the vicinity of the experimental set-up. E.g., the line at 847 keV corresponds to the first excited state in ^{56}Fe, whereas the one at 276 keV corresponds to the first excited state in ^{81}Br present in the detector. The time-of-flight distribution of the latter one is converted into an energy spectrum and used to determine the neutron detection efficiency, as described in Ref. [9]. Below we show results from an experiment on ^{235}U(n$_{th}$,f), obtained by applying this neutron efficiency to data taken at the 10 MW research reactor of the Centre for Energy Research in Budapest.

The prompt fission γ-rays were selected by choosing a narrow TOF window, the background from other reactions was assessed and subtracted, and the obtained energy spectrum was normalized with the number of fission events. In order to deduce the emitted prompt fission γ-ray spectrum, the measured spectrum has to be corrected with the response function of the used detectors, which were determined by means of Monte Carlo simulations with the computer code PENELOPE2011 [12], folded with the energy resolution of the corresponding detector. More detailed information on the actual extraction of the emission spectrum is described e.g. in Ref. [4]. In this manner we obtained PFGS characteristics, i.e. the average γ-ray multiplicity $\overline{\nu}_\gamma$, the average energy per photon ϵ_γ and the total γ-ray energy $E_{\gamma,tot}$, which are presented below.

3 Results

In this section an overview of experimental results is given for our prompt fission neutron and γ-ray measurements performed so far. The results are compared to other experimental and calculated values, where available. The reactions that were investigated are ^{252}Cf(sf), ^{235}U(n$_{th}$,f) and ^{241}Pu(n$_{th}$,f).

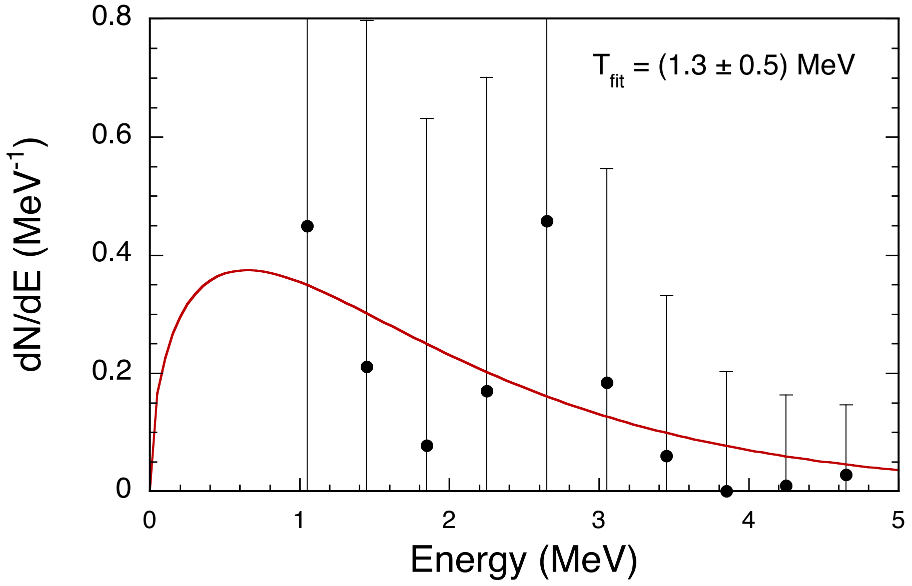

Fig. 2: Energy spectrum of neutrons from the reaction ^{235}U(n$_{th}$, f). The experimental values are fitted to a Maxwell-Boltzmann distribution in order to extract a temperature parameter and the distribution was normalized to 1.

3.1 PFNS from the reaction ^{235}U(n$_{th}$,f)

In an experiment performed at the 10 MW research reactor of the Centre for Energy Research in Budapest, the reaction ^{235}U(n$_{th}$,f) was investigated with the same set-up as used to determine the neutron efficiency mentioned above (see Ref. [10] for details). Fig. 2 shows the obtained neutron energy probability distribution, normalized to one. Due to several experimental problems, leading to the loss of an unknown number of events it was not possible to extract an average number of prompt fission neutrons $\overline{\nu}$. As a consequence of the poor statistics, we had to restrict ourselves to neutron energies above 1 MeV due to uncertainties in assessing the constant background. However, it was at least possible to extend the energy range to 5 MeV. Although the statistical accuracy is quite poor, indicated by the large error bars (containing both statistical uncertainties and those from background determination as well as efficiency), we fitted a Maxwell-Boltzmann distribution to the data. It results in a temperature parameter $T_{fit} = (1.3 \pm 0.5)$ MeV, which agrees well with documented values like e.g. 1.33 MeV from Ref. [11]. Admittedly, the experimental data presented here is of poor quality and the shape of the neutron spectrum - a Maxwell-Boltzmann distribution - was assumed to be known beforehand; still, perfect agreement with previously published values for the temperature parameter was achieved. This result might serve at least to illustrate the applicability of the neutron detection technique mentioned above.

3.2 PFGS characteristics from the reactions ^{252}Cf(sf), ^{235}U(n$_{th}$,f) and ^{241}Pu(n$_{th}$,f)

Several experiments were performed to measure prompt fission γ-ray spectra (PFGS). The investigation of the spontaneous fission of ^{252}Cf was carried out at the DG Research Centre IRMM in Geel, while the thermal neutron-induced fission of both ^{235}U and ^{241}Pu was studied at the 10 MW research reactor of the Centre for Energy Research in Budapest. The experiments were conducted according to the brief description in Sect. 2. More information on the particular instrumentation and experimental set-ups for each measurement as well as details about the data treatment are given in Refs. [4, 13–15], where also the obtained emission spectra are shown. An overview of the determined PFGS characteristics from our measurements, denoted by the individual detector that was used, are displayed in Fig. 3 for ^{252}Cf(sf), in Fig. 4 for ^{235}U(n$_{th}$,f) and in Fig. 5 for ^{241}Pu(n$_{th}$,f). Mean photon multiplicity, mean photon energy per

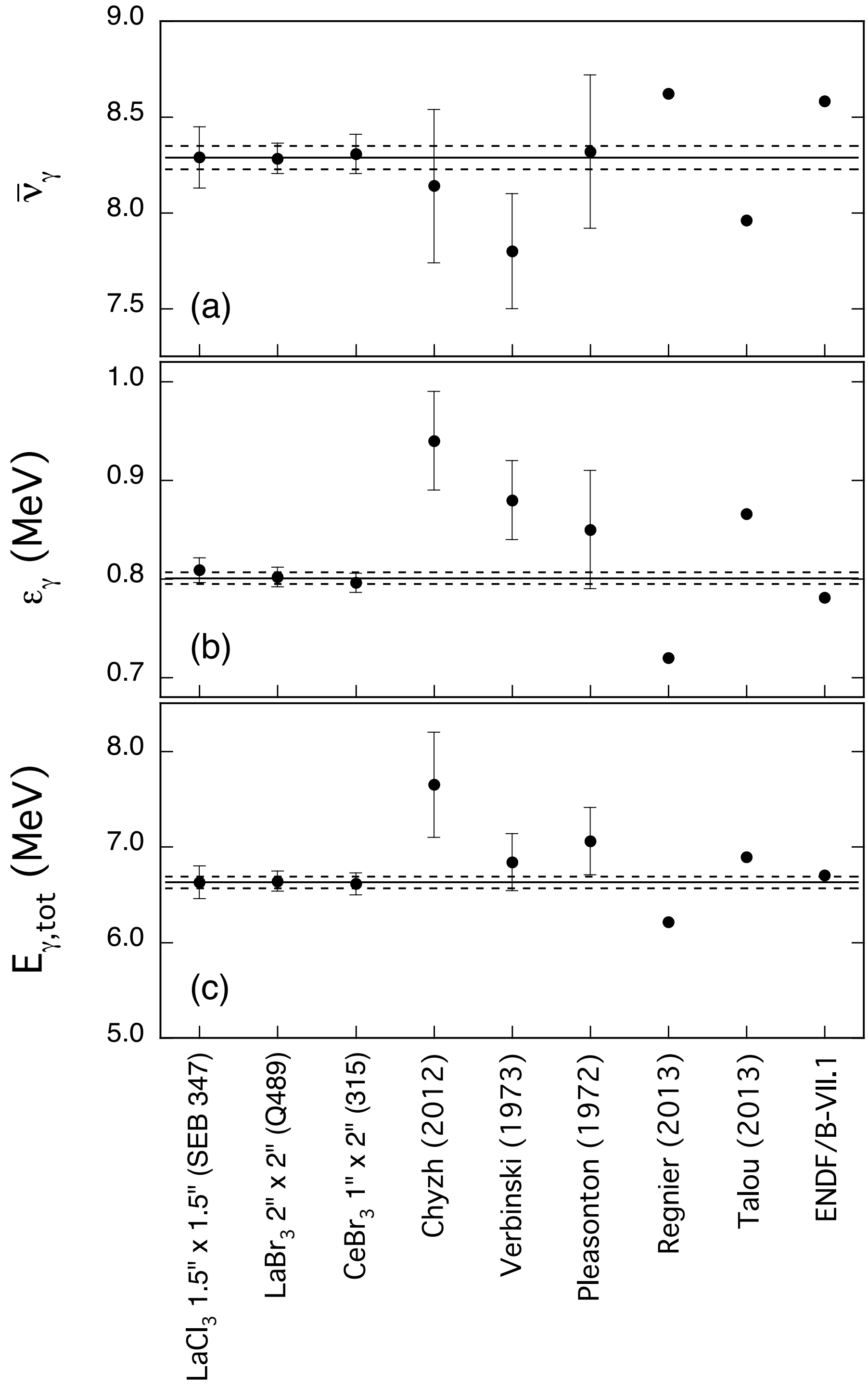

Fig. 3: Overview of measured PFGS characteristics for the spontaneous fission of ^{252}Cf: (a) Mean photon multiplicity, (b) mean photon energy per fission and (c) total released photon energy from our work, denoted by the detectors in use. Average values and their uncertainties are displayed as full drawn and dashed lines, respectively. They are compared to results from other experiments and model calculations (see Refs. [4, 13] and references therein.

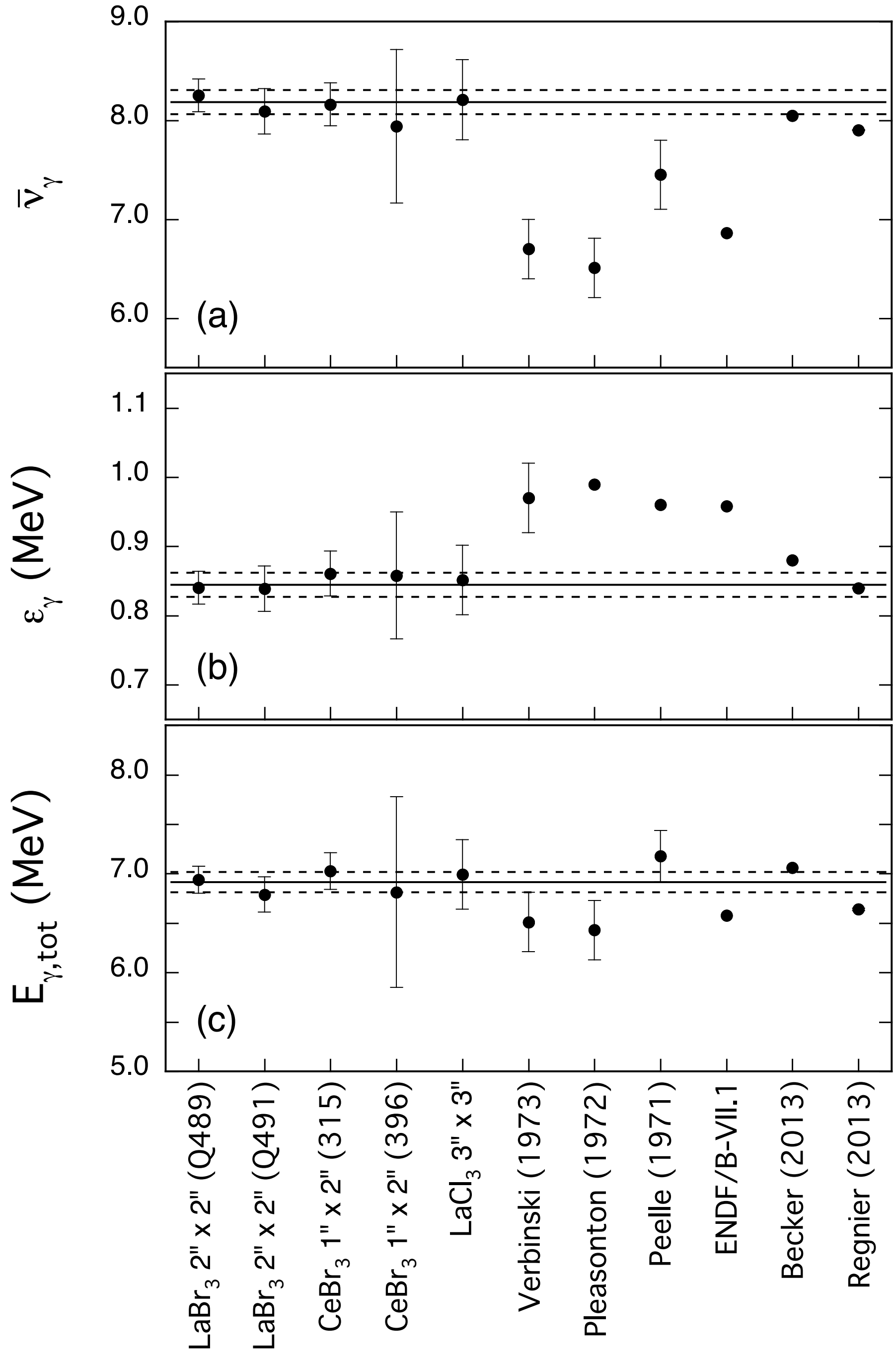

Fig. 4: Overview of measured PFGS characteristics for the thermal neutron-induced fission of ^{235}U: (a) Mean photon multiplicity, (b) mean photon energy per fission and (c) total released photon energy from our work, denoted by the detectors in use. Average values and their uncertainties are displayed as full drawn and dashed lines, respectively. They are compared to results from other experiments and model calculations (see Ref. [14] and references therein.

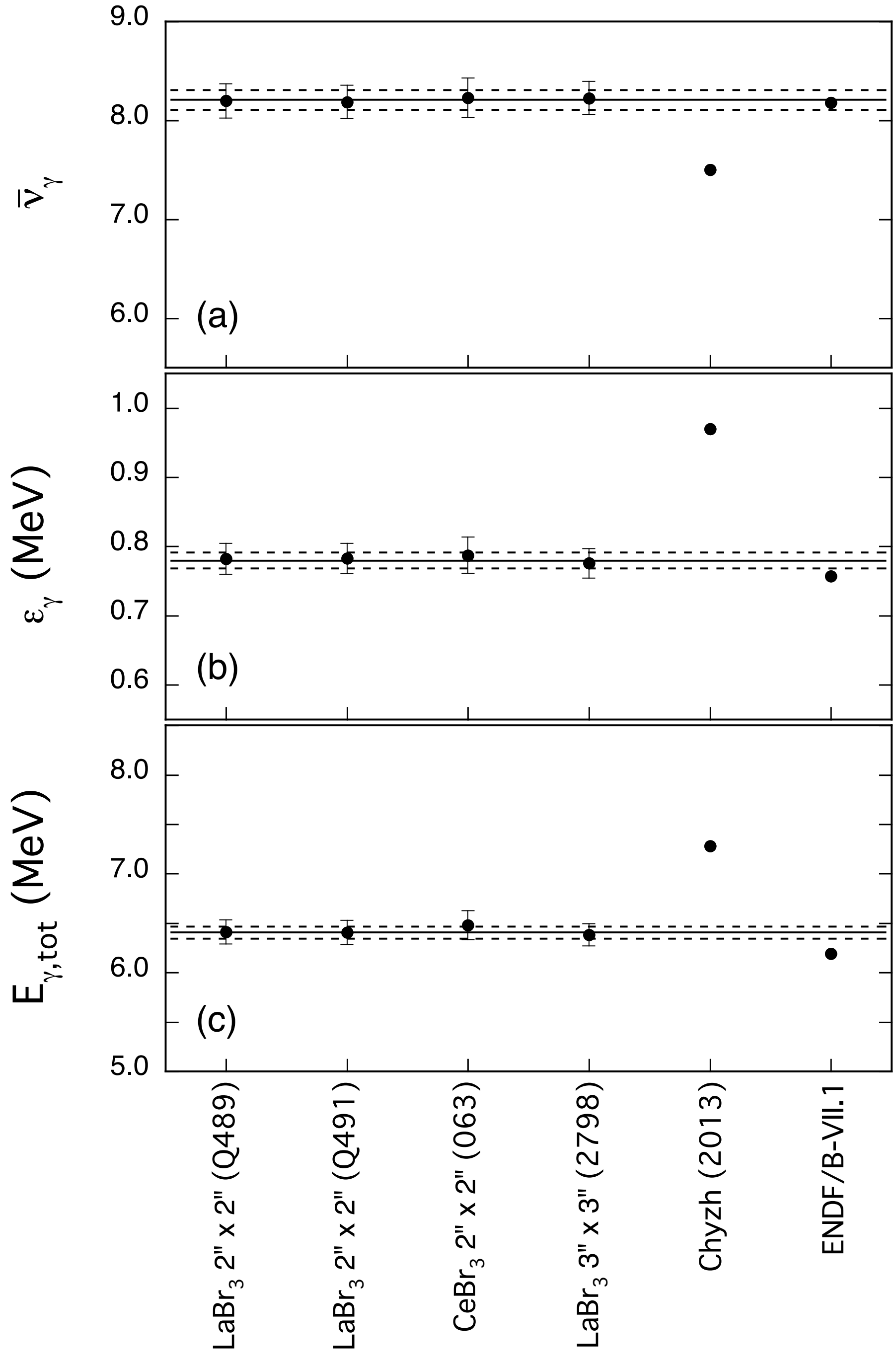

Fig. 5: Overview of measured PFGS characteristics for the thermal neutron-induced fission of ^{241}Pu: (a) Mean photon multiplicity, (b) mean photon energy per fission and (c) total released photon energy from our work, denoted by the detectors in use. Average values and their uncertainties are displayed as full drawn and dashed lines, respectively. They are compared to results from other experiments and model calculations (see Ref. [15] and references therein.

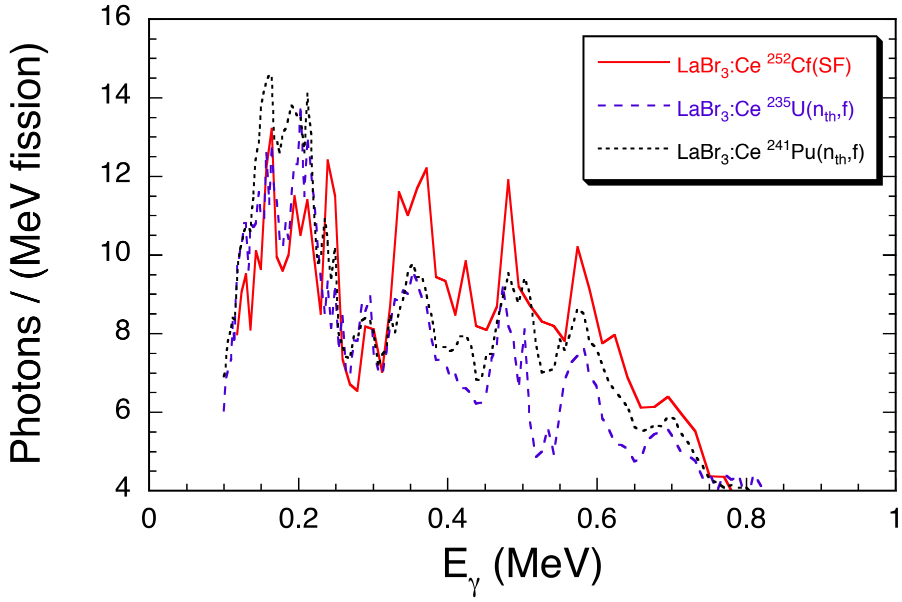

Fig. 6: Low energy part of PFGS for the fissioning systems reported about in this work. The distinct peak structures exhibit obvious similarities.

fission and total released photon energy are plotted in the upper, middle and lower part of the figures, respectively. Values averaged over our results and their uncertainties are shown as as full drawn and dashed lines. Corresponding values from other measurements as well as from theoretical calculations and an evaluated data library are shown for comparison. References to those studies are given in the references mentioned above. A brief discussion of our findings follows below.

4 Summary and discussion

From the overviews presented in the previous section we may conclude that all our measurements gave very consistent results independent of the particular detector in use. That made it possible to determine average values for the PFGS characteristics with hitherto unprecedented accuracy for all fissioning systems investigated so far. Other recent experimental results, taken with the detector system DANCE and published during the years 2012 and 2013, exhibit average multiplicities, which are lower than ours, and a total γ-ray energy released in fission that is too high compared to our results. An explanation for this discrepancy is given by absorption effects for low energy γ-rays as addressed in Ref. [4]. A comparison of the low energy regions of the PFGS for the fissioning systems reported about in this work, exhibits distinct and very similar peak structures (see Fig. 6). We believe that their origin is de-excitation of rotational states in mainly heavy fission fragments. A possible confirmation for that is the topic of our efforts in the next future.

We have also demonstrated a technique to measure PFNS with lanthanum bromide detectors by evaluating γ-rays produced in inelastic neutron scattering off bromine nuclei, even if the obtained data for ^{235}U is affected by very low statistics. Since the energy of the considered excited state is equal to the minimum kinetic energy of the detectable neutrons, the low energy threshold in this work should be in principle at about 280 keV. This is much lower than the 500 and 1000 keV, which is reported for standard neutron detectors. In order to prove that we have to apply this technique to experimental data taken with better statistics, which is in progress.

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References

[1] R. Capote,V. Maslov, E. Bauge, T. Ohsawa, A. Vorobyev, M. Chadwick, S. Oberstedt, Consultants' Meeting on Prompt Fission Neutron Spectra of Major Actinides, Summary Report, IAEA-INDC International Nuclear Data Committee, INDC(NDS)-0541 (January 2009).

[2] Nuclear Data High Priority Request List of the NEA (Req. ID: H.3, H.4), www.oecd-nea.org/dbdata/hprl/hprlview.pl?ID=421 and www.oecd-nea.org/dbdata/hprl/hprlview.pl?ID=422.

[3] A. Oberstedt, S. Oberstedt, R. Billnert, W. Geerts, F.-J. Hambsch, J. Karlsson, Nucl. Instr. and Meth. A668 (2012) 14.

[4] R. Billnert, F.-J. Hambsch, A. Oberstedt, and S. Oberstedt, Phys. Rev. C 87 (2013) 024601.

[5] R. Billnert, S. Oberstedt, E. Andreotti, M. Hult, G. Marissens, and A. Oberstedt, Nucl. Instr. and Meth. A647 (2011) 94.

[6] G. Lutter, M. Hult, R. Billnert, A. Oberstedt, S. Oberstedt, E. Andreotti, G. Marissens, U. Rosengård and F. Tzika, Nucl. Instr. and Meth. A703 (2013) 158.

[7] S. Oberstedt, R. Borcea, T. Brys, Th. Gamboni, W. Geerts, F.-J. Hambsch, A. Oberstedt, and M. Vidali, Nucl. Instr. and Meth. A714 (2013) 31.

[8] S. Oberstedt, R. Borcea, F.-J. Hambsch, Sh. Zeynalov, A. Oberstedt, A. Göök, T. Belgya, Z. Kis, L. Szentmiklosi, K. Takàcs, T. Martinez-Perez, Proceedings of Seminar on Fission VII, Het Pand, Gent, Belgium, May 16-20, 2010, eds. C. Wagemans, J. Wagemans and P. D'hondt, ISBN-13 978-981-4322-73-7 (2010) 207.

[9] A. Oberstedt, R. Billnert, S. Oberstedt, Nucl. Instr. and Meth. A708 (2012) 7.

[10] A. Oberstedt, R. Billnert, S. Oberstedt, Phys. Proc. 31 (2012) 21.

[11] N. Kornilov, F.-J. Hambsch, I. Fabry, S. Oberstedt, T. Belgya, Z. Kis, L. Szentmiklosi and S. Simakov, Nucl. Sci. Eng. 165 (2010) 117.

[12] http://www.oecd-nea.org/tools/abstract/detail/nea-1525.

[13] A. Oberstedt, R. Billnert, F.-J. Hambsch, and S. Oberstedt, to appear in Phys. Rev. C.

[14] A. Oberstedt, T. Belgya, R. Billnert, R. Borcea, T. Bryś, W. Geerts, A. Göök, F.-J. Hambsch, Z. Kis, T. Martinez, S. Oberstedt, L. Szentmiklosi, K. Takàcs, and M. Vidali, Phys. Rev. C87, 051602(R) (2013).

[15] R. Billnert, T. Belgya, T. Bryś, W. Geerts, C. Guerrero, F.-J. Hambsch, Z. Kis, A. Moens, A. Oberstedt, S. Oberstedt, G. Sibbens, L. Szentmiklosi, D. Vanleeuw, and M. Vidali, to appear in Phys. Rev. C.