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Systematics of prompt fission γ-ray spectra characteristics

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Abstract

Systematics from 2001, describing prompt fission γ-ray spectra (PFGS) characteristics as function of mass and atomic number of the fissioning system, has been revisited and parameters have been revised, based on recent experimental results. Although originally expressed for spontaneous and thermal neutron induced fission, validity for fast neutrons was assumed and applied to predict PFGS characteristics for the reaction n + ^{238}U up to incident neutron energies of E_n = 20 MeV. The results from this work are compared with preliminary experimental results.

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1. Introduction

A bit more than a decade ago an evaluation of prompt fission γ-ray spectra (PFGS) was presented, trying to describe the average total γ-ray energy released in fission as well as the average energy per emitted γ-ray as functions of mass and atomic number, A and Z, of the fissioning system (Valentine, 2001). From both parameters, even the average γ-ray multiplicity was deduced. Based on thitherto available experimental data for ^{233}U(n_{th}, f), ^{235}U(n_{th}, f), ^{239}Pu(n_{th}, f) and ^{252}Cf(sf) taken until 1973 [see in the work of (Valentine, 2001) for references], A- and Z-dependencies were found "by trial and error", without any physical significance (Valentine, 2001). Nevertheless, the description given there offers the possibility to estimate average properties of PFGS for fissioning systems, which are difficult or virtually impossible to access experimentally.

However, in recent years the measurement of PFGS has undergone a renaissance, motivated by requests for new precise values especially for γ-ray multiplicities and average photon energy release per fission in the thermal neutron induced fission of ^{235}U and ^{239}Pu (NEA, 2006). Two experimental groups, a collaboration involving IRMM/Chalmers/Budapest and another between Los Alamos/Livermore, reported on results from ^{252}Cf(sf) (Billnert

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et al., 2013a; Oberstedt et al., 2014a) and ^{235}U,^{241}Pu(n$_{th}$, f) (Oberstedt et al., 2013; Billnert et al., 2013b, 2014) as well as ^{252}Cf(sf) (Chyzh et al., 2012, 2013) and ^{235}U(n, f) (Chyzh et al., 2013), ^{239}Pu,^{241}Pu(n, f) (Chyzh et al., 2013; Ullman et al., 2013), respectively. A comparison of all results for the PFGS properties from ^{235}U(n$_{th}$, f) (Oberstedt et al., 2013), ^{241}Pu(n$_{th}$, f) (Billnert et al., 2013b, 2014) and ^{252}Cf(sf) (Oberstedt et al., 2014a) exhibits deviations, which motivates an investigation of the influence of the recent measurements on the evaluation from (Valentine, 2001). This will be done in the following section. Since this evaluation was performed for and, hence, the systematics that is based upon it, may be assumed to be valid for thermal neutron induced an spontaneous fission, the excitation energy of the fissioning system ranges from zero to the neutron separation energy. Below we extend the revised systematics to fast neutron induced fission and apply it to the system n + ^{238}U, which is highly relevant for fast reactor applications and one of the 6 important isotopes in the focus of the CIELO (Collaborative International Evaluated Library Organization) Pilot Project of the OECD/NEA (CIELO, 2013). The predicted PFGS characteristics for this system up to 20 MeV incident neutron energy are then compared to results from theoretical calculations and recent measurements.

2. Revised systematics for PFGS characteristics

According to (Valentine, 2001), the average total γ-ray energy released in fission $E_{\gamma,tot}$ is depending linearly on the prompt fission neutron multiplicity $\overline{\nu}_n$, which is based on the study published by (Nifenecker et al., 1972). However, the latter cited work was extended from the spontaneous fission of ^{252}Cf to other fissioning systems by including a dependence from both their mass and atomic numbers, A and Z, respectively. The suggested description for $E_{\gamma,tot}(\overline{\nu}_n$,Z,A) in MeV is of the form

$$E_{\gamma,tot}(\overline{\nu}_n, Z, A) = \varphi(Z, A) \times \overline{\nu}_n + 4.0 \quad , \tag{1}$$

with

$$\varphi(Z, A) = a_0 + a_1 \times Z^2 A^{1/2} \quad . \tag{2}$$

The parameters a_0 and a_1 were determined by a least-squares fit to experimental data, while the values for $\overline{\nu}_n$ had been taken from experiments (Valentine, 2001). The average energy per emitted γ-ray ϵ_γ was assumed to be independent from $\overline{\nu}_n$ and depending on A and Z according to

$$\epsilon_\gamma(Z, A) = b_0 + b_1 \times Z^{1/3} A^{-1} \quad . \tag{3}$$

Here too, the parameters b_0 and b_1 were determined by a fit to experimental results. A relation for the average prompt fission γ-ray multiplicity $\overline{\nu}_\gamma$ may then be inferred by dividing Eq. 1 with Eq. 3 and using Eq. 2. Although different functions may be used to approximate $\overline{\nu}_\gamma(\overline{\nu}_n$,Z,A), we have chosen

$$\overline{\nu}_\gamma(\overline{\nu}_n, Z, A) = c_0 + c_1 \times \overline{\nu}_n \times Z^{5/3} A^{-1/3} \quad , \tag{4}$$

in order to present experimental values graphically. Figure 1 gives an overview of all experimental results for (a) $E_{\gamma,tot}(\overline{\nu}_n$,Z,A), (b) ϵ_γ(Z,A) and (c) $\overline{\nu}_\gamma(\overline{\nu}_n$,Z,A) in accordance with the above given equations. The full drawn (black) lines correspond to the evaluation by (Valentine, 2001), based on experimental results that were reported until 1973 (for references we refer to (Valentine, 2001)), denoted by full drawn (black) circles. The (blue) open squares indicate the results obtained by the Los Alamos/Livermore collaboration (Chyzh et al., 2012, 2013; Ullman et al., 2013) by using the DANCE detector system (Heil et al., 2001). Our results, also recently published (Billnert et al., 2013a; Oberstedt et al., 2014a, 2013; Billnert et al., 2013b, 2014) are shown as (red) open circles. The values for $\overline{\nu}_n$ were taken as given by (Valentine, 2001). Due to obvious discrepancies between the historical and the recently obtained experimental data, a new evaluation seems to be reasonable on the basis of these new results. However, even those exhibit considerable differences depending on by which experimental group they were obtained. The reason for that has been explained by absorption effects in the DANCE system for γ-rays with energies below 500 keV (Billnert et al., 2013a), leading to lower γ-ray multiplicities and an overestimation of the average total γ-ray energy. Hence, those values are perhaps not quite reliable and therefore not included in a new evaluation, whose result is depicted by (red)

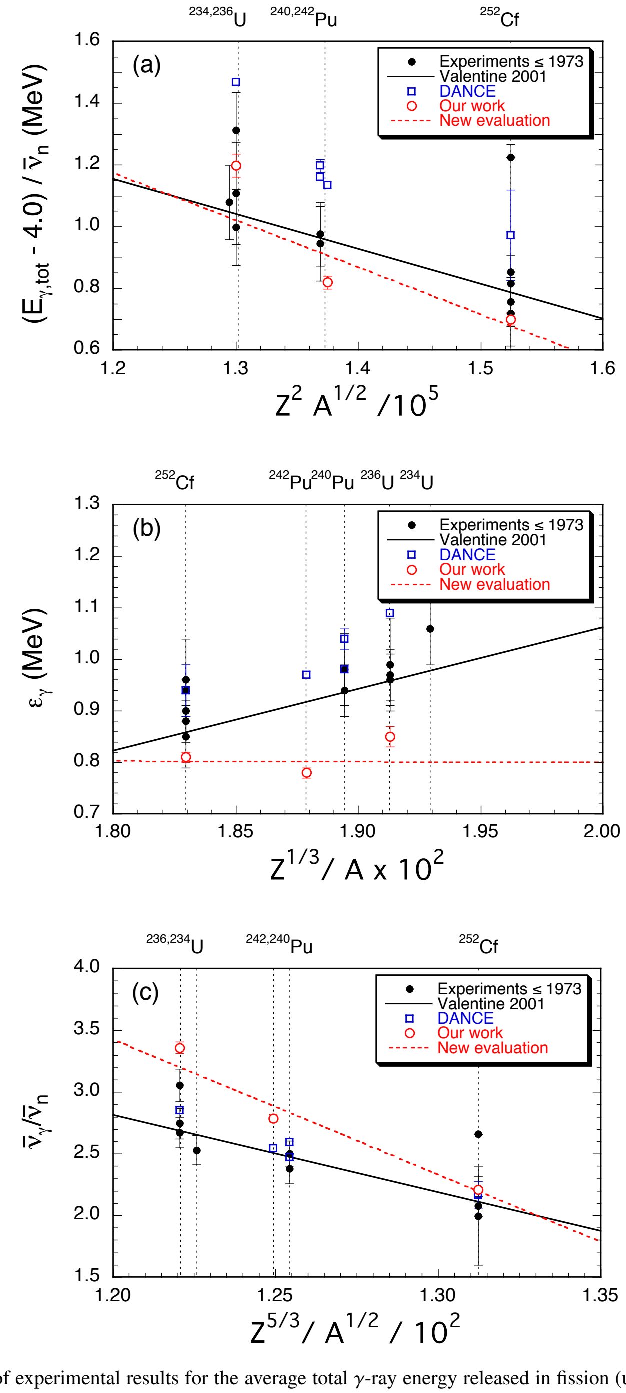

Fig. 1. (Color online) Overview of experimental results for the average total γ-ray energy released in fission (upper part), the average energy per photon (middle part) and prompt fission γ-ray multiplicity (lower part) as function of A and Z for different fissioning systems. Full (black) circles denote historical results (see (Valentine, 2001) for references), open (blue) squares indicate results obtained with DANCE (Chyzh et al., 2012, 2013; Ullman et al., 2013) and open (red) circles represent results from our previous work (Billnert et al., 2013a; Oberstedt et al., 2014a, 2013; Billnert et al., 2013b, 2014). Also shown are results from evaluations by (Valentine, 2001) (solid black line) and from this work (dashed red line), based on the historical data and our results, respectively. For the sake of clarity, the corresponding fissioning systems are given, too.

dashed lines in Fig. 1. They were obtained by least-squares fits, weighted with the uncertainties, to our experimental results for ^{235}U(n$_{th}$, f) (Oberstedt et al., 2013), ^{241}Pu(n$_{th}$, f) (Billnert et al., 2013b, 2014) and ^{252}Cf(sf) (Billnert et al., 2013a; Oberstedt et al., 2014a).

Obviously, the are differences between our evaluation and the one by (Valentine, 2001); in particular, our results predict an average γ-ray energy that is practically the same for all fissioning systems. Of course, this has to be subject for further experimental studies, which then have to be included in the systematics presented here. For the time being, we emanate from the results of our evaluation to predict PFGS properties for the system n + ^{238}U.

3. The system n + ^{238}U at E$_n$ ≤ 20 MeV

From the systematics of PFGS characteristics presented in the previous section it should be possible to interpolate to any fissioning system. The only apparent energy dependence is the one that is hidden in the prompt fission neutron multiplicity. If this one is known, there is no obvious reason why the validity of this systematics should be restricted to spontaneous or thermal neutron induced fission. Hence, in the following we apply the systematics to fission induced by fast neutrons on ^{238}U in the energy range from 0 to 20 MeV. The energy dependence of the prompt fission neutron multiplicity is a linear one, according to both (Madland, 2006) and probably the most recent evaluated data file (ENDF/B-VII.1, 2011). However, in the considered energy range channels for multi-chance fission may be open, leading to the emission of pre-fission neutrons. Since these neutrons are not emitted from fission fragments but the compound system, they don't contribute to the de-excitation of the fragments in competition with prompt γ-ray emission. However, as shown by (Chen and Liu, 2011), they are included in the numbers given in the evaluated files. Hence, pre-fission neutrons have to be assessed and subtracted in order to obtain proper values to be used in the systematics above.

As already shown by (Nifenecker et al., 1972) for ^{252}Cf(sf), the total γ-ray energy released in fission (and the γ-ray multiplicity) is increasing linearly with the average number of neutrons emitted per fission, i.e. $\overline{\nu}_n$. The same behavior may be inferred for ^{235}U(n, f) and ^{238}U(n, f) from (Madland, 2006), where a linear increase of both total γ-ray energy and average prompt neutron multiplicity with incident neutron energy is reported. However, it was shown by (Chen and Liu, 2011) for the neutron induced fission of ^{235}U that this is only true as long as the (n, f) channel is considered. Hence, for neutron energies above the neutron separation energy of the compound system, the channels for second, third etc. fission, i.e. (n, nf), (n, 2nf) and so on, may be open and the neutrons emitted prior to fission of the corresponding residual compound systems have to be subtracted from the total number of prompt fission neutrons. Details about the calculations will be found elsewhere (Oberstedt et al., 2014b); here we have to restrict ourselves to presenting the results of the prediction of PFGS properties.

4. Results and conclusion

From the revised systematics presented in Sect. 2 and the results for the prompt neutron multiplicity from the fission fragments obtained according to the considerations briefly discussed in Sect. 3, PFGS properties were inferred for the fissioning system n + ^{238}U. The results are summarized in Fig. 2. The upper part shows the average total γ-ray energy released in fission as a function of incident neutron energy (Fig. 2(a)). Our result (solid red line) is denoted as prediction and shown together with a linear fit to an empirical approach from (Madland, 2006) (dotted black line) and preliminary experimental results from (Laborie, 2013) (full black circles) for E_n = 1.7 and 15.6 MeV (Laborie et al., 2012). In contrast to our results, the data from (Madland, 2006) do not exhibit any kinks at the thresholds for second and third chance fission, which should appear, if the evaporation of neutrons prior to fission had been corrected for. They appear also in the predictions for the average prompt fission γ-ray multiplicity (middle part) and the average γ-ray energy per fission (lower part), both as function of incident neutron energy as depicted in Fig. 2(b) and (c), respectively. Unfortunately, only very preliminary results from (Laborie, 2013) were available so far for comparison, which however deviate among each other much more than expected. In Fig. 2(c), the (black) dashed line indicates a constant value for the the average γ-ray energy, according to Eq. 3, which contains no $\overline{\nu}_n$ and, hence, no energy dependence. The solid (red) line exhibits kinks and is the result of dividing the total γ-ray energy with the average

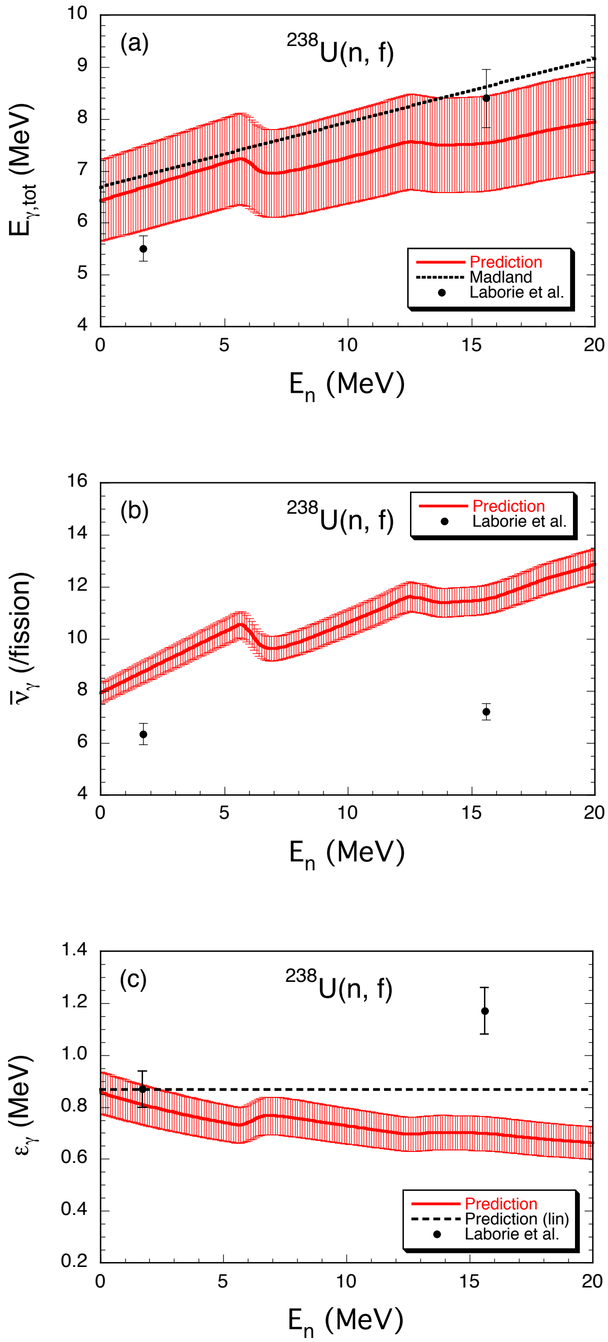

Fig. 2. (Color online) Predicted PFGS characteristics for n + ^{238}U: (upper part) average total γ-ray energy released in fission, (middle part) average prompt fission γ-ray multiplicity, and (lower part) average γ-ray energy per fission, all as function of incident neutron energy. The results from this work are depicted as solid (red) lines and compared to preliminary experimental results from (Laborie, 2013) (full black circles). A linear approximation from (Madland, 2006) for the average total γ-ray energy is shown as dotted (black) line.

prompt fission γ-ray multiplicity. Which description is to prefer here can't be decided from the comparison with the preliminary experimental values from (Laborie, 2013).

We have shown that the systematics, which was originally found for thermal neutron induced and spontaneous fission (Valentine, 2001), may as well be applied to fission induced by fast neutrons as long as the corresponding prompt fission neutron multiplicities are known and correctly used. This implies that pre-fission neutrons must be subtracted from the total average prompt fission multiplicities, when the de-excitation of fission fragments is considered. The results from our predictions, at least with respect to the shape of the curves, are supported by observations for the systems n + ^{232}Th, n + ^{235}U and n + ^{237}Np, as reported by (Fréhaut, 1988). Our predictions build on reasonable assumptions, which in principle may be transferred to any fissioning system. So far they suffer from considerable uncertainties, basically from the fits of the systematics in Sect. 2. However, more reliable experimental data included in the systematics will certainly reduce the uncertainties of the fit parameters and, hence, of the predictions. For the system n + ^{238}U new experimental results are under way. New measurements at E_n = 1.7 MeV (again) and 5.2 MeV are currently being analyzed (Laborie, 2013), and first results for the energy region E_n = 0.7 - 4.0 MeV, from an experiment performed at the novel LICORNE facility of IPN Orsay (Lebois et al., 2014; Wilson et al., 2013), were recently presented (Lebois et al., 2013). Correction factors for the absorption of low energy γ-rays, affecting the data obtained by the Los Alamos/Livermore collaboration with the DANCE detector system, will be presented soon (Oberstedt et al., 2014a), which means that a broader basis will soon be provided for a revised systematics of PFGS properties and thus predictions with higher precision. Also results from new model calculations for the system n + ^{238}U have been reported recently, obtained with the FIFRELIN code (Regnier et al., 2013) for E_n = 1.8 MeV (Litaize et al., 2013) and in the framework of the Point-by-Point model [for details see e.g. (Tudora, 2013a) and references therein], which cover the entire energy range investigated in this work (Tudora, 2013b). These results together with a more detailed description of this work will be shown elsewhere (Oberstedt et al., 2014b).

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