# REACTOR $\overline{\nu}_e$ PROPERTIES FROM $\beta$ -DECAY STUDY OF FISSION FRAGMENTS BY TOTAL ABSORPTION SPECTROSCOPY\*

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Total absorption spectroscopy is a unique technique that is highly efficient for detecting  $\gamma$  radiation in a wide range of energies. Thanks to this feature, total absorption spectrometers are successfully used to study  $\beta$  decay of unstable nuclei, allowing to determine  $\beta$ -feeding distributions over the entire decay energy window. Recent studies show the importance of total absorption spectroscopy measurements of the  $\beta$  decay of fission product for the understanding of the reactor  $\overline{\nu}_e$  spectra. This contribution focuses on the unique capabilities of Modular Total Absorption Spectrometer (MTAS) allowing us to obtain  $\beta$  intensities and the de-excitation path of excited levels in daughter nucleus. These capabilities are applied to the analysis of decay data of <sup>86</sup>Br, abundantly produced in nuclear reactors. MTAS results affecting the shape of the  $\overline{\nu}_e$  spectrum associated with <sup>86</sup>Br decay are presented.

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## 1. Introduction

The nuclear  $\beta$  decay is the most common form of radioactivity of unstable nuclei. Owing to its simple nature, the transformation of one nucleon

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into another of opposite isospin, the decay mechanism and selection rules are well-understood. However, the simple picture gets complicated once we account for the effects related to the configuration mixing in multi-nucleon systems. In addition, just a few nucleons away from stability, the  $\beta$ -decay energy window,  $Q_{\beta}$ , is large enough for transitions populating high-density states above pairing gap to occur. It has been proven that the  $\beta$ -decay schemes obtained from the high-resolution study are burdened with systematic error due to the inability to detect numerous weak  $\gamma$  rays de-exciting highly excited states in the daughter nucleus [1–4].

Accurate measurements of  $\beta$  decays of the most abundantly produced fission fragments are important to improve the reference calculations of fission reactor  $\overline{\nu}_e$  flux. The number of reactor  $\overline{\nu}_e$  interactions with detector matter measured through inverse  $\beta$  decay was found about 5% smaller than the expected number of events. This disparity has been dubbed the "reactor antineutrino anomaly" [5–7]. Reactor antineutrino spectra are obtained from  $\beta$ -spectrum measurements of <sup>235</sup>U, <sup>239</sup>Pu, and <sup>241</sup>Pu thermal fission and <sup>238</sup>U fast fission [8–11]. Integral  $\beta$  distributions are converted to  $\overline{\nu}_e$  spectra by fitting a number of virtual  $\beta$  branches. Thousands of  $\beta$  transitions are replaced with several effective branches. This simplification brings the risk of systematic errors, such as, for example, inaccurate shape of the  $\beta$  spectrum due to the existence of forbidden transitions [7]. An alternative procedure — the summation method — uses decay schemes of all fission products, summing each nuclide's  $\overline{\nu}_e$  spectrum weighted by its fission yield [6, 12]. This approach requires an accurate measurement of individual  $\beta$  decays of fission products. Incomplete information about the decay schemes tends to underestimate the probability of  $\beta$  transitions feeding high-excited states, which artificially shifts the calculated  $\overline{\nu}_e$  flux to higher energies. This causes an overestimation of the predicted number of potentially detected  $\overline{\nu}_e$ .

Total absorption spectroscopy with sodium iodine NaI(Tl) crystals is a very powerful technique to study  $\beta$  decay of unstable nuclei. Thanks to the very high efficiency for detecting  $\gamma$  radiation in a wide range of energies total absorption spectroscopy provides a reliable measurement of the emitted  $\gamma$  rays and resulting  $\beta$ -strength distribution over the entire decay energy window. The Modular Total Absorption Spectrometer (MTAS) is the largest total absorption spectrometer in operation in the world, and it has a superior segmentation. It consists of 19 hexagonal-shaped crystals of 21" length and 8" diameter, arranged in three-ring geometry, see Fig. 1. A detailed description of the detector can be found in Karny *et al.* [13].



Fig. 1. Modular Total Absorption Spectrometer coupled with a silicon strip detector.

Experimental MTAS spectra can be described as the sum of the detector response to  $\beta$  and following  $\gamma$ -ray transitions weighted by the intensity of the individual  $\beta$  transitions, see Eq. (1)

$$\boldsymbol{R} = \sum_{j=0}^{N} b_j \, \boldsymbol{r}_j \,, \tag{1}$$

where  $b_j$  is the intensity of  $\beta$  transition feeding level j,  $r_j$  is the detector response to  $\beta$  and  $\gamma$ -ray transitions corresponding to the level j,  $b_j$  parameters can be obtained by fitting the simulated spectra to the experimental result. Different approaches to fitting algorithms can be used, see, *e.g.* Ref. [14].

The analysis procedure requires precise knowledge of the detector response to  $\gamma$  and  $\beta$  radiation, which can be achieved by modeling the detector using the Geant4 simulation package [13] and its verification measurements with well-known calibration sources. The second requirement is the knowledge of levels de-excitation path. In the case of less exotic nuclei with low Q value, all levels and  $\gamma$ -ray transitions are usually known from highresolution measurements. For more exotic nuclei, most high-energy levels are missed. One way to determine the de-excitation path is to assume E1, M1, and E2  $\gamma$ -ray transitions to all known levels. Spins and parities of new bins can be obtained from the Gamow–Teller selection rules. However, the information of spins and parities for the known part of the level scheme can be incomplete. This requires adopting some assumptions, leading to different possible level schemes for each nuclide. This solution favors high-energy  $\gamma$ -ray transitions, which in the case of wrong estimation of bins spin and parity or incomplete information of the known part of the level scheme can lead to systematic errors that are difficult to estimate or eliminate.

Figure 2 shows an exemplary MTAS response to a fictitious 3 MeV energy level, assuming different possible de-excitation paths: by one 3 MeV  $\gamma$  ray, two  $\gamma$ -ray transitions of energies 1 and 2 MeV, and de-excitation path with both possibilities, each with a probability of 50%. The upper panel presents the results for all MTAS, for which the differences between the responses are not large. The bottom panel presents the spectrum of the MTAS central detector only, which resembles a response to the discussed radiation from a small total absorption spectrometer. As the size of the detector decreases, the knowledge of the de-excitation path becomes increasingly important.

This contribution presents a unique analysis of MTAS data, which allows to obtain levels de-excitation path based on data from  $^{86}$ Br  $\beta$  decay.



Fig. 2. (Color online) Simulated total absorption response to the  $\gamma$  rays emitted from a fictitious 3 MeV level. Solid (blue) curve represents the de-excitation path of one 3 MeV  $\gamma$  ray, dashed (red) curve shows the response to two  $\gamma$  rays of the energies of 1 and 2 MeV. The dotted (yellow) curve represents the de-excitation path consisting in 50% of the single 3 MeV  $\gamma$  ray and in 50% of two  $\gamma$  rays emission. The upper panel shows the response of the total MTAS, the bottom panel that of the central detector.

## 2. Experimental set-up

The measurements were performed at the Holifield Radioactive Ion Beam Facility (HRIBF) at Oak Ridge National Laboratory (ORNL). A 40 MeV proton beam irradiated a <sup>238</sup>UCx target placed inside the ion source. Fission

products were mass selected by means of electromagnetic on-line separation and implanted onto a movable tape. The implantation point was located in front of MTAS, as shown in Fig. 1. The tape transported the radioactive samples into the center of MTAS, between two silicon strip detectors, and removed them after an adjustable measurement time. Such a measurement cycle was repeated till the anticipated statistics was achieved.  $\beta$  decay of over 70 fission products has been studied during several experimental campaigns.

The  $\beta$ -decay spectrum of <sup>86</sup>Br obtained from the MTAS measurement and compared with the simulated detector response assuming the decay scheme published in the ENSDF database is shown in Fig. 3. The gray/light blue area denotes the part of the spectrum that cannot be reproduced by changing the intensities of known  $\beta$  transitions and new levels fed by  $\beta$  decays need to be added.



Fig. 3. (Color online) Experimental total energy deposit in the MTAS detector of the  $\gamma$  and  $\beta$  rays emitted in the decay of <sup>86</sup>Br (solid black) compared with the simulated detector response based on ENSDF data (dashed gray/blue). The gray/light blue area denotes the part of the spectrum that cannot be reproduced by known  $\beta$  transitions.

#### 3. Analysis

The first step of the analysis requires modifications of intensities of known  $\beta$  transitions. In addition, 27 energy bins between  $E_{\min} = 4.8$  MeV and 7.8 MeV were added. The de-excitation paths were determined by assuming E1, M1, and E2  $\gamma$ -ray transitions to the all known levels as described in the previous section. After adjusting  $\beta$ -feeding pattern, the intensities of  $\gamma$ -ray transitions were established and compared with experiment.

To facilitate the analysis of  $\beta - \gamma$  decay pattern, a two-dimensional spectrum of total energy deposit in MTAS plotted on the X-axis and energy deposited in the individual detectors on the Y-axis was prepared. The spectral projection on the X-axis represents the  $\beta$  transitions, while the projection on the Y-axis the individual  $\gamma$ -ray transitions. By placing the gate along the Y-axis to the energy area corresponding to the position of the examined bin, a histogram showing the  $\gamma$ -ray transitions emitted from this level was obtained. An analogous spectrum was determined for the simulation result, and then the probabilities of the  $\gamma$ -ray transitions were modified to obtain agreement between the simulated and the experimental spectrum. The procedure applied for the 100 keV wide energy bin centered at 6.5 MeV is schematically illustrated in Fig. 4.



Fig. 4. (Color online) Two-dimensional spectrum of <sup>86</sup>Br decay (upper right panel). The total energy deposit in MTAS is plotted on the X-axis, while the signals from the external modules counted separately are plotted on the Y-axis. The lower panel shows the 2D spectrum projected on X-axis (black/red line) and simulated detector response for the 6.5 MeV energy bin. Vertical lines mark selected events. The left panel shows the 2D spectrum gated on 6.45–6.55 MeV energy region and projected on Y-axis. The black line shows the experimental histogram, the gray/red line shows the result of the simulation after  $\gamma$ -ray intensities fit.

The presented method allows to obtain coincidence relations and deexcitation path of  $\gamma$ -ray transitions from the individual bins, which is possible due to the highly modular construction of MTAS. However, despite of the MTAS discovery potential, the method has some limitations. The basic limit is the finite possibility to isolate events related to de-excitation path of a specific level due to the limited energy resolution. Second limitation is the statistics. The analysis requires a large number of counts throughout the entire energy range. In the case of low number of events in the bin, like the ones at the high-energy end of the collected spectra, it is possible to find only the strongest  $\gamma$ -ray transitions.

In the analysis of the  $\beta$  decay of <sup>86</sup>Br, the intensities of most  $\gamma$ -ray transitions emitted from new levels to the ground state were overestimated and needed to be corrected following the two-dimensional coincidence spectra, see Fig. 4. The final result of the total energy deposit in MTAS together with the simulation is shown in Fig. 5. In addition, the spectrum from MTAS external modules was plotted, as a supplementary check that the  $\gamma$ -ray intensities were correctly identified, as shown in Fig. 6.



Fig. 5. (Color online) Experimental total energy deposit in MTAS emitted in the decay of <sup>86</sup>Br (gray) compared with the simulated detector response based on the new decay scheme (dashed/red). Other curves represent detector responses to individual levels, including ground state (dark red).  $E_{\min}$  indicates the energy above which new levels have been added.



Fig. 6. (Color online) Experimental energy deposit in external MTAS modules to  $\gamma$  and  $\beta$  transitions emitted in the decay of <sup>86</sup>Br (gray) compared with the simulated detector response based on ENSDF data (dashed black/blue curve) and the final result of the analysis (solid gray/red curve).

### 4. Results

Following the analysis of MTAS data,  $\beta$  transitions feeding low-lying levels in the 2.5–3.0 MeV range were significantly reduced. Over 20% of  $\beta$  feeding was shifted to the levels above 5.5 MeV. The ground state level feeding was increased from 15% up to over 20%. This result agrees with that recently published by Rice *et al.* [15] from a measurement also performed with the total absorption technique. Figure 7 presents the changes in the cumulative intensities of  $\beta$  transitions in comparison with the corresponding ENDSF entry.

The new decay scheme was used to determine the  $\overline{\nu}_e$  energy distribution and the probability of its detection. For this purpose,  $\overline{\nu}_e$  energy spectra were calculated assuming an allowed  $\beta$  shape and zero recoil energy of the daughter nucleus. The average probability of detecting an  $\overline{\nu}_e$  is an integral of a product of the  $\overline{\nu}_e$  spectra and the inverse  $\beta$ -decay cross section [16]. Figure 8 shows the emitted and detected  $\overline{\nu}_e$  energy distribution. The detection probability, expressed in  $10^{-43}$  cm<sup>2</sup> units, changes from 2.62(75) to 2.46(5) with respect to the calculation based on ENSDF entry.



Fig. 7. (Color online) Cumulative feeding of <sup>86</sup>Kr states after <sup>86</sup>Br  $\beta$  decay. Solid (blue) line follows ENSDF data, dashed (red) line corresponds to the presented MTAS data evaluation.



Fig. 8. (Color online)  $\overline{\nu}_e$  energy distribution emitted in the decay of <sup>86</sup>Br calculated based on ENSDF decay scheme (thick solid black/blue line) compared with the results of the MTAS measurements (solid gray/red line). The dashed black curve shows the shape of the inverse  $\beta$ -decay cross section [16]. Thick dashed black/blue and gray/red dashed lines show the product of the  $\overline{\nu}_e$  energy distribution and the inverse  $\beta$ -decay cross section for ENSDF and MTAS data, respectively.

#### 5. Summary

This contribution presents the Modular Total Absorption Spectrometer (MTAS) capabilities to obtain not only  $\beta$  intensities but also the deexcitation path of excited levels in daughter nucleus. MTAS is the largest

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segmented total absorption spectrometer and its properties allow establishing reliable information about the pattern of  $\gamma$ -ray transitions. The described spectrum de-convolution technique was used to analyze <sup>86</sup>Br  $\beta$ -decay data. The new <sup>86</sup>Br decay scheme is different from the current ENSDF entry for this nucleus, while being in a good agreement with the recent total absorption spectroscopy measurement by Rice *et al.* [15]. The decay scheme was used to determine the  $\overline{\nu}_e$  energy distribution and the probability of its detection by inverse  $\beta$ -decay reaction. The  $\overline{\nu}_e$  detection probability become smaller by about 6%, changing from 2.62(75) × 10<sup>-43</sup> cm<sup>2</sup> to 2.46(5) × 10<sup>-43</sup> cm<sup>2</sup>, with respect to the calculation based on ENSDF data.

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