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A decay database of coincident $\gamma-\gamma$ and $\gamma-X$-ray branching ratios for in-field spectroscopy applications

A.M. Hurst[1,*], *B.D.* Pierson[2], *B.C.* Archambault[2], *L.A.* Bernstein[1,3], and *S.M.* Tannous[1]

[1]Department of Nuclear Engineering, University of California, Berkeley, California, 94720, USA
[2]Pacific Northwest National Laboratory, Richland, Washington, 99352, USA
[3]Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California, 94720, USA

Abstract. Current fieldable spectroscopy techniques often use single detector systems heavily impacted by interferences from intense background radiation fields. These effects result in low-confidence measurements that can lead to misinterpretation of the collected spectrum. To help improve interpretation of the fission products and short-lived radionuclides produced in a composite sample, a coincidence-γ database is being developed in support of a robust portable γ and X-ray coincidence detector system concurrently under development at the Pacific Northwest National Laboratory for in-field deployment. Hitherto, no database exists containing coincident $\gamma-\gamma$ and $\gamma-X$-ray branching-ratio intensities on an absolute scale that will greatly enhance isotopic identification for in-field applications. As part of this project, software has been developed to parse all radioactive-decay data sets from the Evaluated Nuclear Structure Data File (ENSDF) archive to enable translation into a more useful JavaScript Object Notation (JSON) formats that more readily supports query-based data manipulation. The coincident database described in this work is the first of its kind and contains coincidence $\gamma-\gamma$ and $\gamma-X$-ray intensities and their corresponding uncertainties, together with auxiliary metadata associated with each decay data set. The new JSON format provides a convenient and portable means of data storage that can be imported into analysis frameworks with relatively low overhead allowing for meaningful comparison with measured data.

1 Introduction

Forensics applications for nuclear security and nonproliferation require accurate information on decay spectroscopy for a large variety of long- and short-lived fission-product debris nuclides. However, the complexity of the spectra collected from in- and near-field measurements makes it difficult to unambiguously identify candidate isotopes and deduce material composition. Indeed, many isotopes contain γ rays at similar energies. And some weaker, yet important, γ-ray lines may be completely obscured by different signatures at nearby energies. Such scenarios are particularly problematic in strong-background environments where isotopic identification from singles decay-spectroscopy measurements becomes increasingly challenging. Coincidence-decay measurements, both $\gamma-\gamma$ and $\gamma-X$ ray, particularly in cases involving low-energy transitions in high-Z material where internal conversion often dominates, can greatly reduce the background and enhance the signal of interest. To help facilitate this need, a new portable γ and X-ray detector system is being developed at the Pacific Northwest National Laboratory (PNNL). A computer-aided design (CAD) rendering of the device is shown in Figure 1, which comprises two cerium bromide ($CeBr_3$) crystals for γ detection, and two silicon-drift detector (SDD) arrays for X-ray detection. Each SDD array comprises a hexagonal arrangement of six *outer* and one *innner* silicon-detector elements. i.e., a total of seven silicon-detector elements per array.

For effective use, this new coincidence-detection device also requires an accurate database of threat signatures. However, the current decay data in the nuclear structure libraries, e.g., the Evaluated Nuclear Structure Data File (ENSDF) [1] and the Live Chart of Nuclides [2], do not provide coincidence $\gamma-\gamma$ and $\gamma-X$-ray intensities needed on an absolute scale, nor any tools to readily deduce this information. To circumvent this problem, we have also developed a coincidence-decay database of $\gamma-\gamma$ and $\gamma-X$-ray data for all coincidence pairs in the residual decay scheme corresponding to the daughter nucleus produced in a radioactive-decay process. The coincidence information includes coincidence γ-ray energies and uncertainties, K-shell X-ray energies and uncertainties in addition to the calculated $\gamma-\gamma$ and $\gamma-X$-ray intensities on an absolute scale in units of percent, together with their associated uncertainties. To accomplish this task and generate the required data, we first developed a parser to convert all of the α-, β^--, and ϵ/β^+-decay data sets from ENSDF into a more useful JavaScript Object Notation (JSON) format. The development of this parser benefits from earlier work to translate ENSDF data sets into an eXtensible Markup Language (XML) format; a very early implementation of this parser is described in a laboratory report in Ref. [3]. In these proceedings, we describe how the required decay

*e-mail: amhurst@berkeley.edu

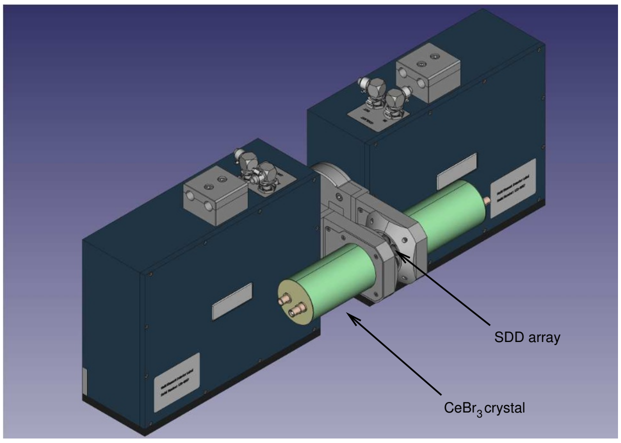

Figure 1. CAD-render of the γ/X-ray coincidence-detector system under development at PNNL.

data are used to develop the coincidence calculations and a few select results are presented.

2 Electromagnetic-transition decay data

Each electromagnetic transition in a given decay scheme can be described by three associated intensity quantities: the γ-ray intensity (Γ_γ); the internal-conversion electron intensity (Γ_e); and the total transition intensity (Γ_T). The total transition intensity may be expressed as

$$\Gamma_T = \Gamma_\gamma + \Gamma_e. \quad (1)$$

Because the total internal-conversion coefficient $\alpha = \Gamma_e/\Gamma_\gamma$, then Γ_T may also be described as the total γ-ray intensity corrected for internal conversion:

$$\Gamma_T = \Gamma_\gamma(1 + \alpha). \quad (2)$$

The values for Γ_γ and α are those parsed from the original ENSDF file, whereupon the α values (where available) were generated using the Band-Raman Internal Conversion Calculator (BRICC) [4] according to the associated transition multipolarity. The corresponding values of Γ_e and Γ_T can then be calculated from the definition of α above, and Equation 2.

In order to calculate coincidence intensities from the γ-ray emission, the aforementioned Γ_γ, Γ_e, and Γ_T quantities must be normalized to the sum of all N total-transition intensities deexciting a given level, such that their branching ratios may be cast as:

$$b_{\gamma_i} = \frac{\Gamma_{\gamma_i}}{\sum_{i=1}^{N} \Gamma_{T_i}}; \quad b_{e_i} = \frac{\Gamma_{e_i}}{\sum_{i=1}^{N} \Gamma_{T_i}}; \quad b_{T_i} = \frac{\Gamma_{T_i}}{\sum_{i=1}^{N} \Gamma_{T_i}}. \quad (3)$$

In cases where there is only a single γ-ray transition ($N = 1$) deexciting a level then $\sum_{i=1}^{N} \Gamma_{T_i} = 1$.

Because it is the strongest γ- and X-ray transitions that provide the useful signatures in forensics applications, in this first version of the database we are only concerned with X rays originating from the K shell. In most cases the K-shell X-rays provide the strongest contribution to the observed X-ray spectrum. The associated K-shell conversion-electron intensity may be written as

$$\Gamma_{e_K} = \Gamma_\gamma \alpha_K, \quad (4)$$

where α_K is the K-shell conversion coefficient at a particular γ-ray transition energy (E_γ). Provided that E_γ exceeds the K-shell binding-energy threshold, i.e., $E_\gamma > E_K$, then the K_α X-ray intensity contribution from each K-shell converted transition may be stated as

$$I_{X_{K_{\alpha_i}}} = f_{K_{\alpha_i}} \Gamma_{e_K}, \quad (5)$$

where $i = 1$, 2, or 3, and $f_{K_{\alpha_i}}$ is the corresponding fluorescence-yield corrected X-ray intensity branching ratio for the K_{α_i} X-ray transition. Similarly, the individual K_β X-ray intensity contributions for each transition are given as

$$I_{X_{K_{\beta_i}}} = f_{K_{\beta_i}} \Gamma_{e_K}, \quad (6)$$

and once again $i = 1$, 2, or 3. All X-ray branching ratios are taken from the *"Table of Isotopes"* [5].

In the case of α and β^- decay, the total-projection X-ray intensity for any particular X-ray transition is simply given as the sum of all contributions from transitions that exceed the subshell binding-energy threshold. For example, the total X-ray intensity for the K_{α_1} line is given by

$$I^{\rm tot}_{X_{K_{\alpha_1}}} = f_{K_{\alpha_1}} \sum_{j=1}^{M} \Gamma_{e_K}, \quad (7)$$

where M represents the total number of transitions that satisfy $E_\gamma > E_K$. Similar expressions may be adopted for all $I^{\rm tot}_{X_{K_{\alpha_i}}}$ and $I^{\rm tot}_{X_{K_{\beta_i}}}$ X-ray lines observed in the total-projection spectra for α- and β^--decay radionuclides. In ϵ/β^+ decay, the fraction of K-shell electron capture to individual

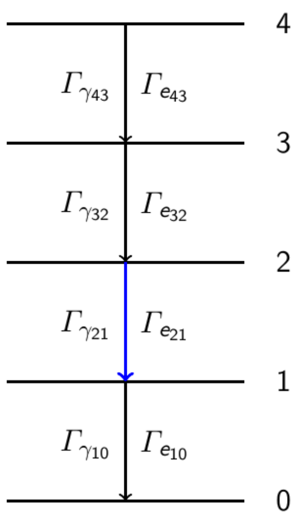

Figure 2. Hypothetical decay scheme used to illustrate coincidence relationships by imposing a gate corresponding to the $2 \to 1$ γ-ray transition (blue arrow). All quantities are described in Sect. 2.

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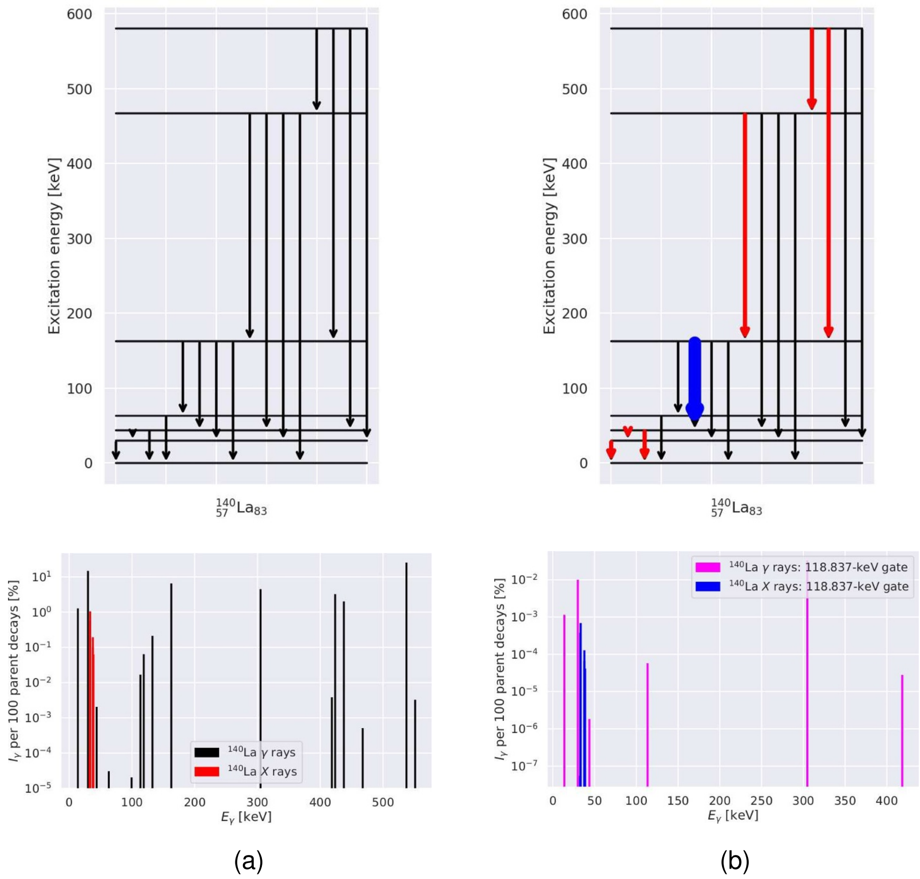

Figure 3. The upper panel in (a) reveals the levels and gammas observed in the decay scheme of the daughter nucleus ^{140}La following the β^- decay of ^{140}Ba [7], while that of (b) also shows those γ rays in red that are in coincidence with the blue γ-ray gating transition at 118.8 keV. The spectra below each respective decay scheme corresponds to (a) the total-projection intensities of all γ rays and X rays, and (b) the expected calculated gated-projection of γ rays and X rays in coincidence with the 118.8-keV γ-ray transition. The presented spectra have not been corrected for any detector-efficiency or photon-attenuation effects.

levels also needs to be accounted for and combined with Equation 7 to explain the observed intensity in the total-projection spectrum.

3 Coincidence-decay intensity relationships

Consider the simple hypothetical decay scheme shown in Figure 2, where all γ-ray transitions are in coincidence with one another. Using the $2 \rightarrow 1$ transition as an example gating transition and the relationships described in Sect. 2, we can calculate the expected coincidence intensities between the γ_{21} transition with all other transitions as

$$\begin{aligned} I_{\gamma_{21}-\gamma_{10}} &= b_{\gamma_{10}}\Gamma_{\gamma_{21}}, \\ I_{\gamma_{21}-\gamma_{32}} &= b_{\gamma_{21}}\Gamma_{\gamma_{32}}, \\ I_{\gamma_{21}-\gamma_{43}} &= b_{\gamma_{21}}b_{T_{32}}\Gamma_{\gamma_{43}}. \end{aligned} \quad (8)$$

Here, b_γ and b_T are the normalized γ-ray and total-transition branching ratios, respectively, and Γ_γ is the absolute γ-ray intensity. Similar expressions based on these relationships can be extended to describe more complicated decay schemes and analogous considerations can be adopted to describe $\gamma - e_K$ coincidences needed to calculate $\gamma - X$-ray coincidence intensities. In decay schemes where there exists parallel paths between a pair of coincident transitions, the individual contributions from independent cascade paths must first be determined and then combined to give the correct coincidence intensity.

4 Results

We have developed a Python-coded translator and parsed over 3200 data sets from ENSDF, encompassing all α-, β^--, and ϵ/β^+-decay data sets, and converted them into a representative JSON format. These translated data sets contain all information from the primary and continuation records of the corresponding ENSDF file [6] serialized into data structures of objects and arrays. In addition to the explicit information parsed directly from the ENSDF data set, these new JSON data structures contain additional information implicit from the decay, e.g., all level energies are now indexed, final energy levels are associated with all reported γ rays, all spin-parity (J^π) permutations for a particular level are deduced and stored in arrays. These new JSON structures provide a far more convenient means for further data processing and manipulation. Accordingly, we have developed an algorithm based on the methods outlined in Sects. 2 and 3 to calculate coincidence intensities using deserialized γ-ray information from the associated ENSDF-translated JSON data structures. Approximately 75% of these ENSDF-translated JSON data structures contain γ-ray intensity information that is then used to create a new set of coincidence-decay JSON data structures. Of the remaining $\sim$ 25%, α decay represents the largest contingent of data sets containing no γ-ray information.

A new schema has been developed describing the quantities stored in the processed coincidence-decay JSON structures based on an intuitive syntax. Although the principal information contained therein are the coincidence γ-ray energy and intensity pairs, together with associated level scheme information (level indices, energies and half lives), we also store additional information including singles γ and X-ray energies and intensities, normalized γ, conversion-electron, and total-transition intensity branching ratios along with the fluorescence-yield corrected X-ray branching ratios from the Table of Isotopes [5]. Furthermore, these data structures contain valence electronic configurations and the electron-subshell binding energies of Ref. [4]. The new coincidence-decay JSON data structures allow for straightforward deserialization of the data in a language-independent manner, hence allowing for users to develop their own automation procedures for access to the data.

To illustrate utilization of the coincidence data, in Figure 3 we present the results for ^{140}La which is populated in the β^- decay of ^{140}Ba ($T_{1/2}$ = 12.751 d) [7], a fission-product nucleus with appreciable cumulative fission yields from several actinides, e.g., thermal (6.314%), fast (5.959%), and 14-MeV (5.6%) neutron-induced fission of ^{235}U [8]. The decay scheme of Figure 3a shows all levels and γ rays observed in the daughter nucleus following ^{140}Ba β^- decay, and the corresponding energy spectrum below shows the total projection of absolute intensities of all γ rays and the calculated X-ray intensities extracted from the appropriate coincidence-decay JSON structure. The impact of coincidence gating is clear from Figure 3b whereby a gating transition imposed on the γ ray at 118.8 keV (162.66 $\rightarrow$ 43.81), indicated by the blue arrow in the decay scheme, reveals six γ rays in coincidence with this gate, indicated by the red arrows in the decay scheme, at: 13.846 keV (43.81 $\rightarrow$ 29.97); 29.966 keV (29.97 $\rightarrow$ 0.0); 43.8 keV (43.81 $\rightarrow$ 0.0); 113.51 keV (581.07 $\rightarrow$ 467.54); 304.85 (467.54 $\rightarrow$ 162.66); 418.44 keV (581.07 $\rightarrow$ 162.66). The expected gated-projection of coincident γ rays and X rays along with their calculated coincident intensities is shown in the lower panel. All data presented in Figure 3 are extracted from the deserialized JSON objects.

5 Summary and outlook

A first of its kind database has been developed containing coincidence-decay energies and absolute intensities (in percentage units), together with their associated uncertainties, from pairs of $\gamma - \gamma$ and $\gamma - X$-ray coincidences observed in radioactive-decay data sets. The coincidence data sets are represented in an open standard JSON format making it easy for applications using different programming languages running in different environments to read and access the data. Additionally, and in order to accomplish this task, we have developed a translator written in Python to convert all radioactive decay data sets into a JSON format. The database files, comprising both the ENSDF-translated JSON data sets as well as the derived coincidence-decay JSON data sets, are currently undergoing testing at PNNL for use in conjunction with the portable γ and X-ray coincidence-detection system concurrently under development. We are also developing an open-source Python package designed to interact with the JSON data structures that we aim to deploy in the near future.

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