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On the need for precise nuclear structure data for high quality $(n, n'\gamma)$ cross-section measurements

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Abstract. The necessary improvement of evaluated nuclear data for nuclear applications development is possible through new and high-quality measurements, often combined with appropriate nuclear-reaction modelling. In particular, improving inelastic cross-section evaluations requires new and high-quality data. We measure $(n, n'\gamma)$ cross-sections using prompt γ -ray spectroscopy and neutron energy determination by time-of-flight. To extract, from these partial data, the total inelastic cross-section, we rely on theoretical model as well as nuclear structure data such as γ ray emission probabilities. This structure information, tabulated in databases, comes with uncertainty. This directly affects the precision of our results, regardless of how good the measurement is. In this paper, we will present the issue of limited precision structure data and its impact on nuclear reaction data quality in the case of neutron inelastic scattering measurements. We will also discuss how to foresee and mitigate the issue.

1 Context

Most nuclear reactor developments rely on evaluated databases for numerical simulations to optimize and predict performance and reactor control parameters. However, these databases still present large uncertainties, preventing calculations from reaching the required precision. An improvement of evaluated databases requires new measurements and better theoretical descriptions of involved reactions. Among the reactions of interest, inelastic neutron scattering (or (n, xn)) are of great importance for the operation of a reactor as they modify the neutron spectrum, the neutron population, and produce radioactive species.

It has been shown that, for example, the current uncertainty on inelastic neutrons scattering off ^{238}U is the dominant source of uncertainty when computing the k_{eff} of a Gas-cooled Fast Reactor (see more in [1]). Accordingly, the measure of $^{238}\text{U}(n, xn')$ is present in the High Priority Request List (HPRL [2]).

To extract, from experimental data, a cross-section value (for example), we usually rely on theoretical model as well as nuclear structure data such as gamma ray emission probabilities. This structure information, tabulated in databases, comes with uncertainty, which will directly affects the precision of experimentally derived results, re-

gardless of how precise the measurement may be. Lowering the uncertainties on nuclear structure data is, therefore, an important step to reach the extra precision expected for the evaluations.

2 Method of measurements

The CNRS-IPHC group is running an experimental program with the GRAPhEME [3] setup installed at the neutron beam facility EC/JRC-GELINA [4, 5] (Geel, Belgium) to measure $(n, xn\gamma)$ reaction cross-sections using prompt γ -ray spectroscopy and neutron energy determination by time-of-flight [6–8]. The obtained exclusive experimental data provide strong constraints on nuclear reaction mechanisms models. So far, data for isotopes such as ^{232}Th [9], ^{235}U [10], ^{238}U [11], $^{182,184,186}\text{W}$ [12] have been recorded and analyzed.

Once the exclusive $(n, xn'\gamma)$ cross-sections are measured, we rely on model calculations (such as TALYS [13], EMPIRE [14] or CoH3 [15]) to find the total (n, n') cross-sections [7]. It is important to remember that these reaction codes use a nuclear level structure description (levels' excitation energy, spin and parities, list of γ transitions and their relative intensities, ...) as input.

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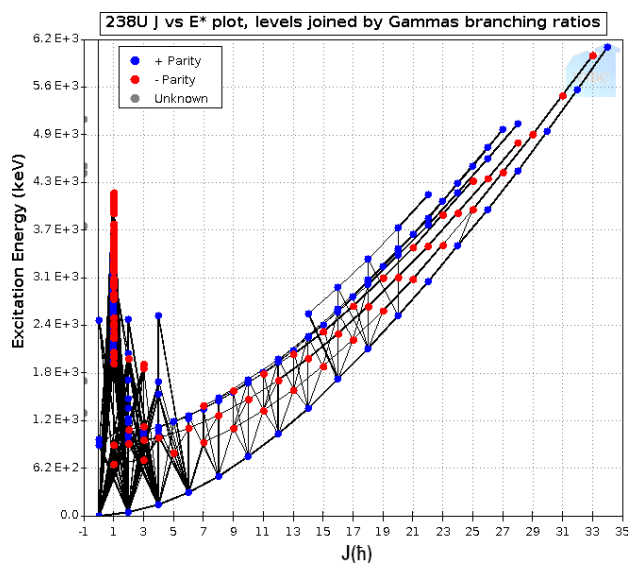


Figure 1. Evaluated states and transitions between them in the ENSDF [16] database for ^{238}U , represented in the J, E^* plane, using the NNDC website tool [19].

3 Flaws in the structure databases.

In the existing databases, such as ENSDF [16] or RIPL-3 [17], the information naturally present some uncertainties (although, in some cases, the uncertainty may not even be known or quoted in the tables¹). But they may also contain some inaccurate information. It is important to point out that this is not due to shortcomings of the maintaining teams or the evaluators filling in the tables, but rather to the lack of experimental data to have final accurate and precise values. In addition, we note that there exists a *bottleneck* of sorts, as it takes time and expertise to move forward from one new experimental value to its integration into the evaluated structure database (and later on in reaction code inputs)

Among these flaws, the first is the *incompleteness* of the evaluated level scheme. Figure 1 shows the representation of the ^{238}U level scheme according to the current values [16, 18] in the J, E^* plane by a dedicated tool [19]. One sees that states at intermediate spin values ($\approx 3 - 9 \hbar$) and higher excitation energy ($> 1.2 \text{ MeV}$) are missing. Additionally, still for the case of ^{238}U , the RIPL-3 [17] database indicates that the level scheme is considered "complete" only up to the 45th level (at 1.4 MeV). Even worse, doubts on spin and/or parity of levels start to appear above level number 10 (at 927 keV). Indeed, it's no surprise, since spectroscopy measurements rely on detectors that often have limited detection efficiency at very high and very low γ energy, leading to missed transitions in the experimental data sets.

These defaults are not specific to ^{238}U . In our work, we identified similar issues for ^{232}Th [21], ^{233}U , ^{239}Pu , ... where we observed some transitions not listed in the evaluated level scheme, as well as some with discrepancies

¹As an example, the ENSDF file for ^{238}U gives the relative intensities of the two γ rays decaying from the 4^+ level at 1056 keV without uncertainty

between the observed branching ratios compared to reference values

The issues in databases are, almost automatically, transposed into models / reaction codes's input files. The Talys [13] manual explicitly indicates that, when it comes to importing a level scheme into the code, "The default choice is the RIPL-3 database(...). Unknown spins, parities and branching ratios are always assigned a value, based on simple statistical spin rules." Because a reaction code cannot accept an empty value or a range, it is natural to make a choice when the original database does not provide a value for a branching ratio (for example). But that may lead to puzzling cases where transitions between two states have an intensity of 0, or three transitions decaying from a level will have each a branching ratio of exactly one third. In consequence, uncertainties in the level databases translate into biases in reaction code. Furthermore, these issues in the input file of codes can persist for some time even if the structure database have fixed the issue.

4 Sensitivity of $(n, n'\gamma)$ cross-sections to structure data

We used a random sampling method to study the sensitivity of calculated $(n, n'\gamma)$ cross-sections to transitions branching ratios. The method is described in another contribution to this conference [22]. It computes the relative standard deviation expected on a calculated $^{238}\text{U}(n, n'\gamma)$ cross-section per relative standard deviation (i.e. uncertainty) on a specific γ transition branching ratio in the level scheme. The resulting sensitivity matrix for transitions in the $^{238}\text{U}(n, n'\gamma)$ reaction (See [23] for full results) is shown in figure 2.

There are two spots with the largest sensitivity (about 0.4 % per %). The first one corresponds to the L13L04² and L13L03 to L04L03 (highlighted in orange in figure 2). This makes sense that if the branching ratios changes between the two transitions from level 13, the cross-section for the 4th to 3rd level will be affected. In fact, the ENSDF file for ^{238}U does not give a branching ratio for the two transitions from the 13th level : L13L03 is listed with an intensity of 100 and 14 uncertainty, and no value (not even a limit) is given for L14L04. Talys (in the version 1.96) uses 100 for L14L03 and 7 for L14L04. The ratio of intensity respects the given uncertainty for L14L03, but it is not known why the value (approx. one half of the uncertainty) has been chosen. The second spot with high sensitivity correspond to the transitions L18L03 and L18L02 to L03L02 (highlighted in green in figure 2). Again, it makes sense the L03L02 cross-section will be sensitive to competing transitions decaying to involved levels. Level 18 is actually the one mentioned in ¹ with relative intensity quoted in the ENSDF file, but no uncertainty. Here, Talys level structure input file uses the same relative intensities that the ones in ENSDF. Similar work of analyzing the sensitivity matrix is done for all blocks of noticeable sensitivity in that manner.

²We are using the Talys notation for transitions. L_iL_j designates the transition from level i and to level j (counting up from the ground state)

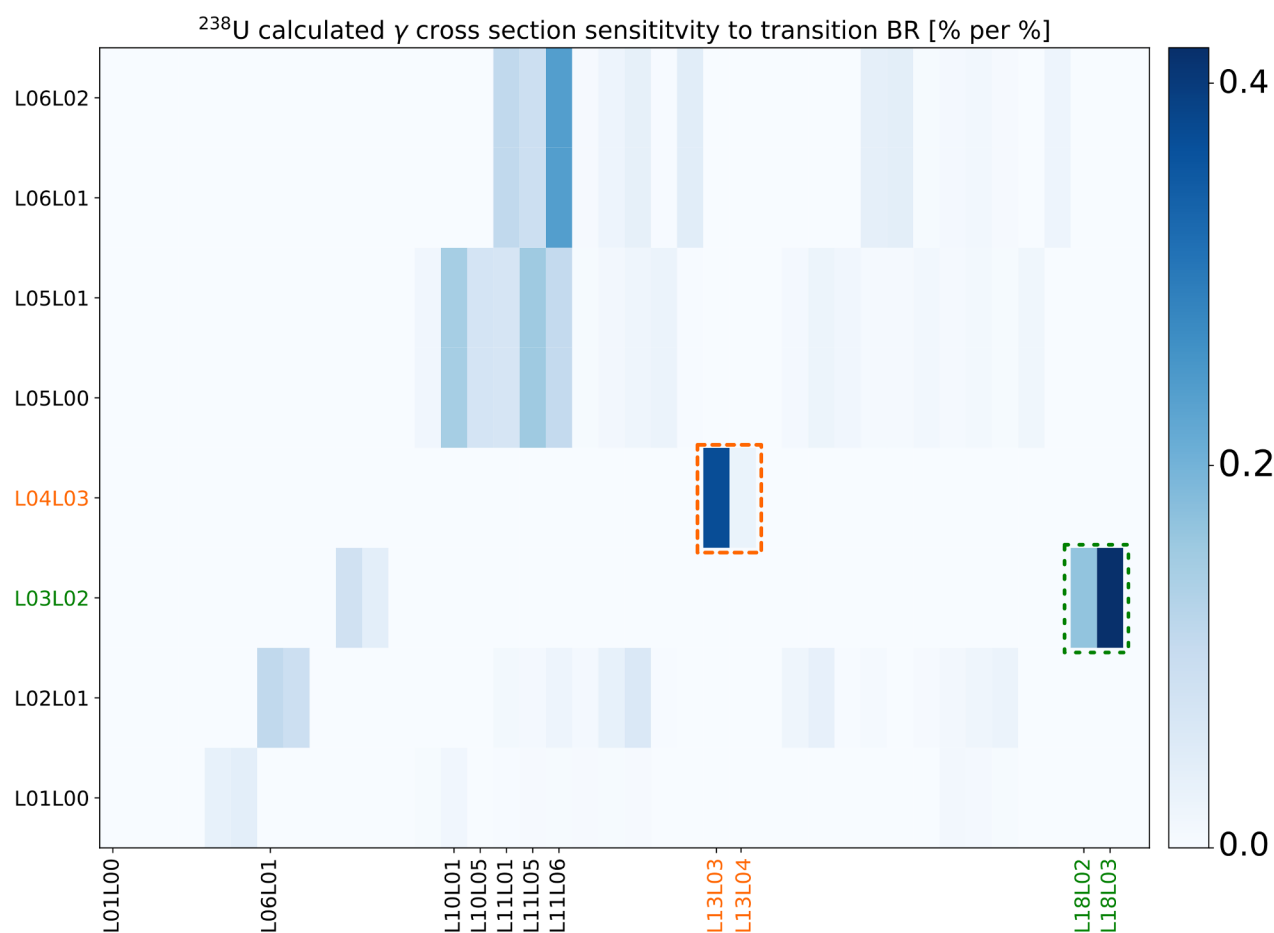


Figure 2. Sensitivity matrix for transitions in the $^{238}\text{U}(n, n' \gamma)$ reaction, in % of cross-section variation per % of branching ratio variation. (Reproduced from [23].) The dashed frames highlight the high sensitivity blocks discussed in the paper.

In general, the sensitivity matrix can be used to check if mismatch between model calculations and experimental value may be attributed to uncertainty on branching ratios. It is also a tool to pinpoint which transitions intensity should have its uncertainty reduced by new measurement or evaluation because of high sensitivity of some cross-section to it.

5 Perspectives

The first action to take in order to tackle the issue of nuclear structure uncertainty for nuclear data production is to raise awareness about it. This is done through communications in conferences, papers and workshop. We also plan to publish our experimental data and their interpretation alongside the nuclear structure data available at the time. This will make any reinterpretation easier if and when updated structure information is made available. The study of sensitivity of calculation codes to the structure input is also a way to ensure the lowest possible dependence of our results to structure issues. The acquisition of new, more precise structure data is a key element. Papers like this one may be used as arguments when presenting an experiment proposal to a selection or funding committee. We can also note that some already existing experimental data, from

experiments not focused on the acquisition of structure information, may be re-examined to extract updated level or transition information, as a "bonus" from the original purpose of the experiment.

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