Towards improvement of the ^{238}U level scheme using γ-spectroscopy of the (n, n'γ) reaction

Carole Chatel[1*], *Jonathan* Wilson[2], *Philippe* Dessagne[1], *Greg* Henning[1] and *Maëlle* Kerveno[1]

[1]Université de Strasbourg, CNRS, IPHC/DRS UMR 7178, 23 Rue du Loess, F-67037 Strasbourg, France
[2]CNRS, IJClab Orsay, bât 100, 15 rue G. Clemenceau, 91406 Orsay Cedex, France

Abstract. Better knowledge of the neutron population in reactors is crucial to improve the accuracy of neutronics simulations of present day or future reactor cores. This population is partially driven by (n, xn) reactions. However, the cross sections of these reactions are not precisely known. This is particularly true for the ^{238}U(n, n'γ) cross section, which is on the High Priority Request List. One method to measure this cross section is to use the prompt γ-ray spectroscopy coupled to time-of-flight measurements. This allows the total (n, n') cross section to be inferred from the measured (n, xnγ) cross sections and the level scheme information. However, the knowledge of the ^{238}U level scheme is still very incomplete, so an initiative to experimentally revisit the ^{238}U structure has been launched using γ-γ coincidences spectroscopy. The ν-Ball γ-spectrometer was coupled to the LICORNE directional neutron source of the ALTO facility, allowing study of inelastic scattering on ^{238}U via γ-γ coincidences. The analysis of data obtained during the first ν-Ball campaign was performed using the Radware escl8r software. At the present time, 73 γ-transitions and 50 levels registered in ENSDF have been confirmed and 120 new γ-transitions and 50 new levels have been found.

1 Introduction

To improve the accuracy of neutronics simulations of present and future reactor cores, a better knowledge of the neutron population is required. This population is, among others, driven by (n, xn) reactions, including inelastic scattering, which change the number of neutrons and their speed. However, their cross sections are, still nowadays, not sufficiently well known. The ^{238}U nucleus is the fertile component of the reactor fuel and constitutes around 90-95% of the fuel mass in present and future cores. Hence, the neutron inelastic scattering cross section of ^{238}U features in the High Priority Request List from OECD/NEA [1], with a target accuracy of 5%, or even down to 2%. One method to obtain this cross section is to use prompt γ-ray spectroscopy coupled to neutron time-of-flight measurements. This allows, from the measured (n, xnγ) cross sections and the level scheme information, to

---

* Corresponding author: c.chatel@gsi.de

infer the total (n, n') cross section [2]. The results of the measured (n,n'γ) cross sections can then also be used in reaction codes which take level scheme information as input, including branching ratios, to calculate (n,xnγ) cross sections. More generally reaction analysis often depend on the knowledge of the nuclear structure of the isotope of interest. The more accurate the results required, the more critical this aspect becomes.

In the case of the ^{238}U, the knowledge of the level scheme is still incomplete: the discrete states are assumed to be fully known up to 1.318 MeV only [3] and the average uncertainties on branching ratios in ENSDF [4] are 8%. Measurements of the (n, n'γ) cross section by the γ-prompt spectroscopy have highlighted gaps in the nuclear structure knowledge and the sensitivity of the cross section to this lack of complete information has been quantified with sensitivity calculations performed with the TALYS code [5]. These show that modifying the branching ratios of 10% in the input's code can have an impact of up to 4% on (n, n' γ) cross sections [2].

For these reasons, improving the ^{238}U level scheme knowledge, even if not exhaustively, has become of high importance. In previous experiments, knowledge of which states are populated by inelastic scattering of fast neutrons on ^{238}U were carried out with only a single Ge detector [6]. To confirm conclusively that a γ-line belongs to ^{238}U requires a double-γ coincidence, which requires a high-efficiency Ge spectrometer and directional high fast neutron fluxes to avoid harming the sensitive Ge detectors. Hence, an initiative to experimentally reinvestigate the ^{238}U nucleus structure has been launched with the γ-γ coincidences method. This can now be done thanks to the coupling between the ν-Ball γ-spectrometer [7] and the LICORNE directional neutron source [8, 9] of the ALTO facility.

The analysis of the γ-γ coincidences matrix obtained during the first ν-Ball campaign with a neutron flux of a mean energy of 1.9 MeV was recently performed using the Radware escl8r software [10]. In this contribution, we will first present the ν-Ball experimental campaign and then present the methodology and the tools used to analyze the γ-γ coincidences matrix. Finally, we will highlight the new results obtained.

2 The ν-Ball experimental campaigns

The two ν-Ball campaigns have been carried out in 2018 and 2022 at the ALTO facility. Within the framework of these two campaigns, two experiments were aiming at studying the structure and determine lifetimes of excited states in exotic neutron-rich fission fragments, by γ-spectroscopy and fast timing technique [11]. Producing such exotic neutron-rich nuclei is possible by fissioning ^{238}U, which produces the highest available N/Z ratio of the fragments after neutron evaporation.

To reduce backgrounds from time-uncorrelated γ-rays (e.g. fragment beta decays), the LICORNE neutron beam is pulsed and only prompt γ-rays from the target are selected. LICORNE produces a pulsed (400 ns period) quasi-mono-energetic kinematically focused neutron flux thanks to the p(^{7}Li, n)^{7}Be inverse reaction. The neutrons impinged on the ^{238}U target with γ-rays emitted from fission products but also from the ^{238}U via the (n, n'γ) reaction. The ν-Ball γ-spectrometer comprised of the two rings of 12 HPGe-Clover detectors and one ring of 10 co-axial HPGe, each surrounded by BGO, and an additional 20 $LaBr_3$(Ce) Fatima detectors [12]. The full energy peak detection efficiency of around 5% at 1 MeV allows for detection of large numbers of γ-γ coincidences. High mass, low-density ^{238}U and ^{232}Th targets were used with naturally collimated neutron beams of average energies of 1.9 MeV and 3.3 MeV. The ν-Ball2 campaign has involved significant technical improvements over the first ν-Ball campaign. The Table 1 summarizes the differences.

Table 1. Differences between the ν-Ball and the ν-Ball2 campaigns.

Campaign	ν-Ball			ν-Ball2
Target	^{238}U		^{232}Th	^{238}U
Neutron energy	1.9 MeV	3.3 MeV	1.9 MeV	1.9 MeV
Avg. Li beam intensity	10 nA	100 nA	80 nA	100 nA
H-gas cell pressure	1.4 atm			1.6 atm
Beam stopper	Pb	Au		
Detectors	24 clover, 10 HPGe, 20 $LaBr_3(Ce)$			24 clover, 20 $LaBr_3(Ce)$
Target shape	81g in 5 disks (2 cm x 1 mm)		131g Th disks in conic Al capsule	71.9 g of ^{nat}U metal turnings in conic Al capsule
Days of data	9	5	19	23

3 Data analysis

3.1 Data analyzed

The ν-Ball2 campaign took place in June and October 2022 with even built data only becoming available in spring 2023. Currently, the analysis of the ^{238}U level scheme has therefore undertaken using data from the first ν-Ball campaign. The data set obtained with an average neutron beam energy of 1.9 MeV have more statistics and less background. A contamination linked to the oxidation of the beam stopper in these data was identified and the problem was corrected for the data taken at 3.3 MeV. In the present work, only the data at 1.9 MeV have been analyzed. Since the aim is to obtain new levels and transitions the energy resolution is privileged and only the data collected by HPGe detectors have been used in the analysis.

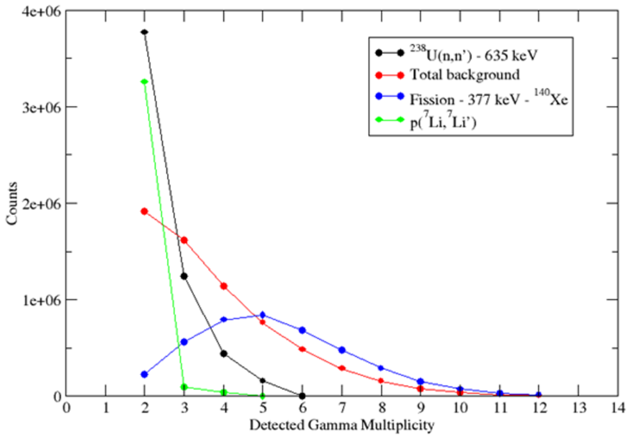

Fig. 1. Number of counts as a function of the multiplicity of the γ-rays coincidence for different reactions.

Finally, conditions on the multiplicity of the coincidences, i.e. the number of γ-rays detected in coincidence in an event are made to reduce the background from fission as much as possible. The detected γ-ray multiplicity distributions from various nuclear reactions occurring have been studied, with results are shown in Fig 1. It can be seen that the reaction (n, n'γ) has a much lower γ-multiplicity than the (n, f) reaction. Thus, considering only a multiplicity of 2 or 3 allows to have a higher inelastic reaction selectivity and to ignore most of the γ-rays coming from fission products. To maximise the signal to background ratio, a condition of multiplicity 2 was initially used.

3.2 Methodology

To realize the analysis, the Radware escl8r software [10] was used. This software has been specially developed to construct complex level schemes from γ-γ coincidence matrices. It allows gates to be placed on selected gamma rays to display all the γ-rays detected in coincidence. A trial level scheme can be constructed and the software will then perform a least-squares global fit to reproduce the associated experimental spectra and coincidence intensities, which can then be improved by iteration. This fit includes the determination of level energies and of intensities of transitions and considers the electron conversion coefficients, the detection efficiency and γ-ray energy calibrations. The Radware escl8r software can also performs with background subtraction.

Since it is assumed that the level scheme is well-known and complete up to 1.318 MeV [3], it was considered that all the γ-rays below 1 MeV and those deexciting the most important levels between 1 and 1.3 MeV (i.e. levels 1037.25 MeV, 1059.66 MeV, 1060.27 MeV, 1105.71, 1128.84 MeV, 1168,88 and 1223,78 MeV) can be used as trustworthy γ-rays to base further analysis on. Hence, gates were set on the deexciting γ-rays of a chosen level, either one after the other (see Fig.2 left, X_{1a} then X_2), either together (see Fig.2 left, X_{1a} and X_2), giving only the common coincidences. The most intense coincidences, and then the weakest ones, were analyzed.

A gate is set on the γ-ray Y to analyze. A first verification is done with ENSDF to check whether this γ-transition is already assumed to belong to the ^{238}U or not, and, if yes, where it is placed. According to ENSDF, another analysis by Govor *et al.* [6] using the (n, n'γ) reaction has been realized to produce the ^{238}U level scheme, but this was only using a single HPGe detector and did not use γ-ray coincidences. In this publication, all the γ-rays that have been detected have been identified. Thus, this publication has been used to check to which nucleus the γ-ray was associated with, and if it was to ^{238}U, to which level. A contamination analysis has also been performed to prevent as much as possible from identifying a γ-ray to belong to the ^{238}U while it belongs to a contaminant. Finally, these ν-Ball data have already been analyzed independently with another method by A. Messingschlager [13]. The results, which have not been published, have also been checked to identify to which level each γ-transition was associated. Thanks to a) these three bibliographic sources plus b) the X_i γ-rays being at the origin of the γ-ray Y analyzed, c) the coincidences with γ-rays Z and X_i that were supposed to be found and d) the contaminations that were listed and compared one by one to the coincidences of the spectrum, the analysis could be performed as follows:

As the structure of such a heavy nucleus is quite complex, a lot of different branches can deexcite a higher energy levels to the ground state. This makes it difficult to observe coincidences of two transitions that are not directly connected. Moreover, restricting the multiplicity to only 2 biases against the branching ratios from cascades containing more than two γ-rays. Thus, if the γ-ray Y in coincidence with X_i belongs to the ^{238}U, Y is either feeding the level that the transition X_i depopulates, either the opposite. In the case of a gate on two γ-rays deexciting the same level a (i.e. X_{1a} and X_2), if the γ-ray in coincidence with both X_{1a} and X_2 belongs to the ^{238}U, this γ-ray feeds the level a. Because of the mass of the target, γ-rays at low energy are significantly attenuated so it is assumed that all the γ-rays below

100 keV are too attenuated or internally converted to be detected. In the special case of an energy difference between two levels below 100 keV (i.e. $E_Y \leq 100$ keV), Y cannot be detected and thus only the γ-rays Z and X_i will appear in coincidence.

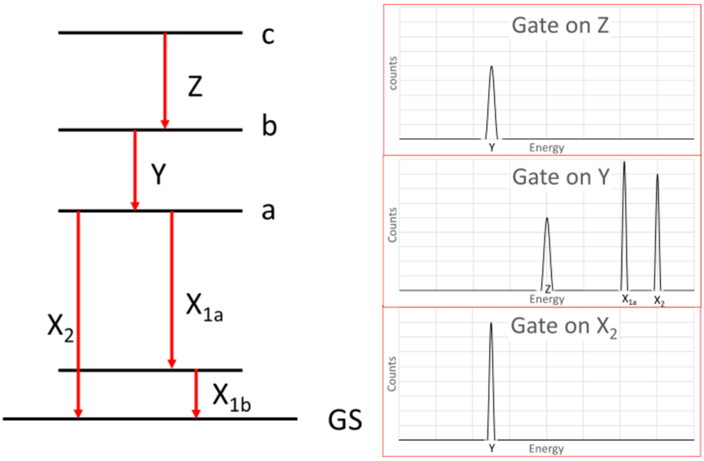

Fig. 2. Level scheme (left) and spectra with gates (right) to schematize Radware escl8r operation.

If the main coincidences were found, the γ-transition was then placed on the level scheme accordingly to the results of ENSDF or Govor *et al.* If the coincidences with all the γ-rays deexciting a level under 1.3 MeV were found, the γ-transition was also placed feeding this level. As the multiplicity condition is 2, no γ-transitions were considered to feed indirectly this level. As mentioned above, the only exception is if two levels have a difference of energy lower than 100 keV. In this case, a γ-ray linking them could exist, which we will discuss later for the 1105.7 keV and 1059.7 keV levels.

3.3 Contamination

During the analysis, most of the contaminations have been identified. These γ-rays come from parasitic reactions of the ^{7}Li beam on the ^{12}C, ^{14}N, ^{16}O and ^{17}Al or from the scattering of neutrons on these same nuclei or on the detectors themselves (Germanium, Bismuth and Oxygen). A small part of the contamination is also due to the fission products; which are also in coincidences with their fission fragment partners. In fact, most of the fission products have been suppressed in the data by choosing a multiplicity of 2 condition. However, the nuclei with the highest fission yields (i.e. ^{140}Xe, $^{132, 134, 136}$Te, ^{102}Zr and ^{96}Sr) are still visible.

Apart from the 511 keV annihilation γ-ray, in total, contaminations from γ-rays deexciting 25 different nuclei have been identified. In addition to the 6 fission products previously cited, those nuclei are $^{70, 72, 74, 76}$Ge from neutron scattering on the Ge detectors and the BGO; ^{209}Bi and $^{17, 18}$O from neutron scattering on the BGO; $^{206, 208}$Pb from neutron scattering on the beam-stopper; ^{14}N from neutron scattering on the air, ^{19}F from neutron scattering on Teflon; ^{27}Al from neutron scattering on the target disks; ^{29}Si due to the reaction of the ^{7}Li-beam on ^{27}Al; ^{18}F and ^{20}Ne due to the reaction of the ^{7}Li-beam on ^{12}C; $^{21, 22}$Ne and ^{22}Na due to the reaction of the ^{7}Li-beam on ^{16}O created by oxidation of the lead present in the beam-stopper; and ^{7}Li, due to deexcitation of the beam-nuclei. This γ-ray has 3 different Doppler-shifted energies corresponding to the 3 different detectors rings.

This identified contamination has been considered during the analysis by created secondary level schemes for all these nuclei in Radware. Thus, Radware escl8r takes them into account when performing the least-squares global fit. The only coincidences between γ-rays that couldn't be considered were those due to the Doppler effect of the ^{7}Li and the fission partners coincidences.

The inventory of these contaminations can be used as a reference for the analysis of later ν-Ball or ν-Ball2 data. Indeed, the contaminations linked to the reactions of the beam on the ^{27}Al and ^{16}O due to Pb oxidation have mostly been suppressed during the ν-Ball2 campaign but all the other ones are expected to occur again. Moreover, three new contaminating γ-rays have appeared due to the presence of gold instead of lead in the beam-stopper.

4 Results

After placing the 50 γ-transitions of the low-lying levels judged as trustworthy in the literature – which have been verified afterwards too – around 150 γ-rays have been analyzed one by one until the present. In total, 100 levels and 193 γ-transitions have been entered in the level scheme. Among the levels, 50 were already written in ENSDF and 50 are new, all of them but 2 being between 1280 keV and 2100 keV, the two others having an energy between 1100 keV and 1250 keV. Among the γ-transitions, 73 were already written in ENSDF and 120 are new. Among those new γ-transitions, 36 are populating or depopulating levels already known in ENSDF and 84 are feeding or depopulating new levels. This analysis is still ongoing.

Because of the target density, γ-rays with an energy lower than 100 keV could not reach the detectors of the ν-Ball spectrometer. However, this study leads us to strongly suspect the existence of a new γ-transition of 45 keV linking the levels 1105.7 keV and 1059.7 keV. It has been decided to place this γ-transition here after noticing that the majority of the γ-transitions feeding the level 1105.7 keV (i.e. γ-rays in coincidence with the γ-rays 957.8 keV and 1060.3 keV) were also in coincidence with the γ-rays deexciting the level 1059.7 keV (i.e. 911.3 keV and 1015 keV). The choice was either to consider all of these γ-transitions as double, i.e. feeding both levels independently, which would have induced a lot of new levels; either to consider a new γ-transition between these two levels. The choice that the 45 keV feeds the level 1059.7 keV and not 1060.3 keV (for which the deexciting γ-rays are 911.9 keV, 1015.3 keV and 1060.3 keV) seemed to be the best according to Radware's fitting but the other solution is still under investigation.

While checking the coincidences, some γ-transitions written in ENSDF have not been found. The most important one is the γ supposed to depopulate the low-lying level at 930.6 keV to the ground state, for which no coincidence has been found. No coincidences have been found for 5 other γ-transitions written in ENSDF. A further analysis will determine if coincidences for each γ-transition written in ENSDF and not entered yet in this level scheme are found.

5 Conclusion and perspectives

Data from the ν-Ball campaign have been analyzed to produce the level scheme of the ^{238}U. This analysis has been performed by using coincidence events with a multiplicity 2 with the Radware escl8r software. Most of the contamination of the spectra has been identified and can be used for the analysis of the ν-Ball2 data.

The ^{238}U level scheme has then been produced until 2625 keV. Until the present, 193 γ-transitions have been identified, populating or depopulating 100 levels. Among them, 50 levels and 120 γ-transitions are not written in ENSDF. Moreover, no coincidences have been found for 6 γ-transitions written in ENSDF, which seem then not to belong to the ^{238}U.

Once this analysis is finished, the coincidences for all the γ-transitions written in ENSDF and not entered yet in the level scheme will be checked. Then, once the level scheme is complete, according to this work, a matrix with a higher multiplicity will be used to precisely determine the branching ratios of the γ-rays. Finally, an analysis will be realized according to the J^π of the final levels to deduce the J^π of the initial level that are not known yet and thus the transition type. It will be also checked whether some new levels found in this work can complete the incomplete bands written in ENSDF. Further information on the J^π could also be deduced by analyzing the angular correlations of the coincidences. Finally, the new levels and γ-transitions will be tested with TALYS to reproduce (n, n'γ) cross sections.

References

1. OECD-NEA, Nuclear data high priority request list, online: http://www.nea.fr/dbdata/hprl/
2. M. Kerveno *et al.*, *Measurement of ^{238}U(n, n'γ) cross section data and their impact on reaction models*, Phys. Rev. C 104, 044605, 2021
3. R. Capote, M. Herman, P. Obložinský, P. Young, S. Goriely, *et al.*, Nucl. Data Sheets 110, 3107, Special Issue on Nuclear Reaction Data, 2009
4. ENSDF: https://www.nndc.bnl.gov/ensdf/
5. A. Koning, S. Hilaire, S. Goriely, TALYS-1.95 A nuclear reaction program, User Manual, 2019
6. L. I. Govor, A. M. Demidov, V. A. Kurkin, and I. V. Mikhailov, *Investigation of 238U in the (n, n'γ) Reaction*, Physics of Atomic Nuclei 77, Issue 2, pp.131-166, 2014
7. M. Lebois *et al.*, *The ν-Ball γ-spectrometer*, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, Volume 960, 21 April 2020, 163580
8. M. Lebois *et al.*, Development of a kinematically focused neutron source with the p(^{7}Li, n)^{7}Be inverse reaction, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, Volume 735, 21 January 2014, Pages 145-151
9. J. N. Wilson *et al.*, *The LICORNE neutron source and measurements of prompt γ-rays emitted in fission*, Phys. Proc. 64, 107−113, 2015
10. Radford, D. C. *ESCL8R and LEVIT8R: software for interactive graphical analysis of HPGe coincidence data sets*, Nucl. Instrum. Meth. Phys. Res. A 361, 297-305, 1995
11. J.-M. Régis *et al.*, *The mirror symmetric centroid difference method for picosecond lifetime measurements via gamma-gamma coincidences using very fast LaBr3(Ce) scintillator detector*, Nuclear Inst. and Meth. in Phys. Research, A 622, 83–92, 2010
12. M. Rudigier, Zs. Podolyák, P.H. Regan *et al.*, *FATIMA — FAst TIMing Array for DESPEC at FAIR*, Nuclear Inst. and Meth. in Phys. Research, A 969, 163967, 2020
13. A. Messingschlager, *Inelastische Neutronenstreuung an ^{238}U mit dem ν-Ball Spektrometer*, Bachelor thesis (private communication), 2020