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Measurement of (n, $xn\gamma$) reaction cross sections in W isotopes

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Abstract. Evaluated nuclear data bases currently used for numerical simulation for the development of nuclear reactors still present large uncertainties. Their improvement is necessary, in particular through better reaction models and nuclear data. Among the reactions of interest, (n, xn) reactions are of great importance for the operation of a reactor as they modify the neutron spectrum, the neutron population, and produce radioactive species. Experimental data on (n, $xn\gamma$) reaction provide strong constraints on nuclear reaction mechanism theories. Tungsten isotopes - which are deformed like actinides but do not fission - are of interest to test the models. ^{182,184,186}W(n, $xn\gamma$) cross sections are measured; results are compared with model calculations by TALYS, EMPIRE and CoH codes.

1. Introduction and motivation

Most nuclear reactor developments today use evaluated databases for numerical simulations to optimize performances and reactor control parameters. However, these databases still present large uncertainties, preventing calculations from reaching high precision. The necessary improvement of evaluations entails new measurements and a better theoretical description of involved reactions [1,2]. Among those, (n, xn) reactions are of great importance for the operation of reactors as they modify the neutron spectrum and produce radioactive species. The group at IPHC started a program to study (n, $xn\gamma$) reactions on actinides in 2005 and already worked on ^{235,238}U and ^{232}Th [3–6], while measurements for other elements were performed and are being analyzed, including ^{nat}W. Tungsten is not an active element in nuclear reactors, but, because of its chemical and mechanical properties [7], it is used in many alloys. The interaction of neutrons with W is therefore of importance for reactor physics. From a theoretical point of view, a better description of (n, xn) reactions on tungsten nuclei allows an improvement of models for other key nuclei in reactors fuel. Indeed, W isotopes are deformed like actinides [8], but easier to describe as they do not present a neutron-induced fission channel. Still, there are only a few measurements available today to test evaluations [9,10]. The experimental data presented here will provide a constraining test of model capabilities for inelastic neutron scattering reactions.

2. Experimental setup

Our measurements are performed at the GELINA facility of the EC-JRC Geel (Belgium). This facility, dedicated to neutron beam experiments, provides a white neutron source with energies from the eV to about 20 MeV along several flight paths with distance of flights up to 400 m. Our experimental setup GeRmanium array for Actinides PrEcise MEasurements (GRAPhEME) uses prompt γ-ray spectroscopy to identify transitions in the nucleus of interest, with the energy of the incoming neutron determined via time of flight. It is located 30 meters away from the neutron source. The setup comprises 4 planar HPGe detectors surrounding the irradiated sample. The incident neutron flux is measured by a fission chamber ahead of the sample. The whole setup is equipped with a digital acquisition. The ratio of detected γ rays for a given transition to the number of neutron leads to the cross section for the transition. A more detailed description of the experimental setup and method is given in Refs. [11] and [12]. In order to produce the most complete data set, measurements were performed with ^{nat}W and isotopically enriched ^{182,184,186}W targets. This will allow precise cross-checks and normalization between isotopes. The ^{nat}W target had a thickness of 0.2 mm, or 0.385 g/cm^2. The isotopic targets were enriched to 90% of the isotope of interest, with thickness from 1.1 to 1.4 g/cm^2 and a density of 10.6 g/cm^3.

3. Experimental results

Some preliminary results of the data analysis for ^{182,184,186}W are shown in Fig. 1 for transitions in the fundamental rotational band and Fig. 2 for transitions

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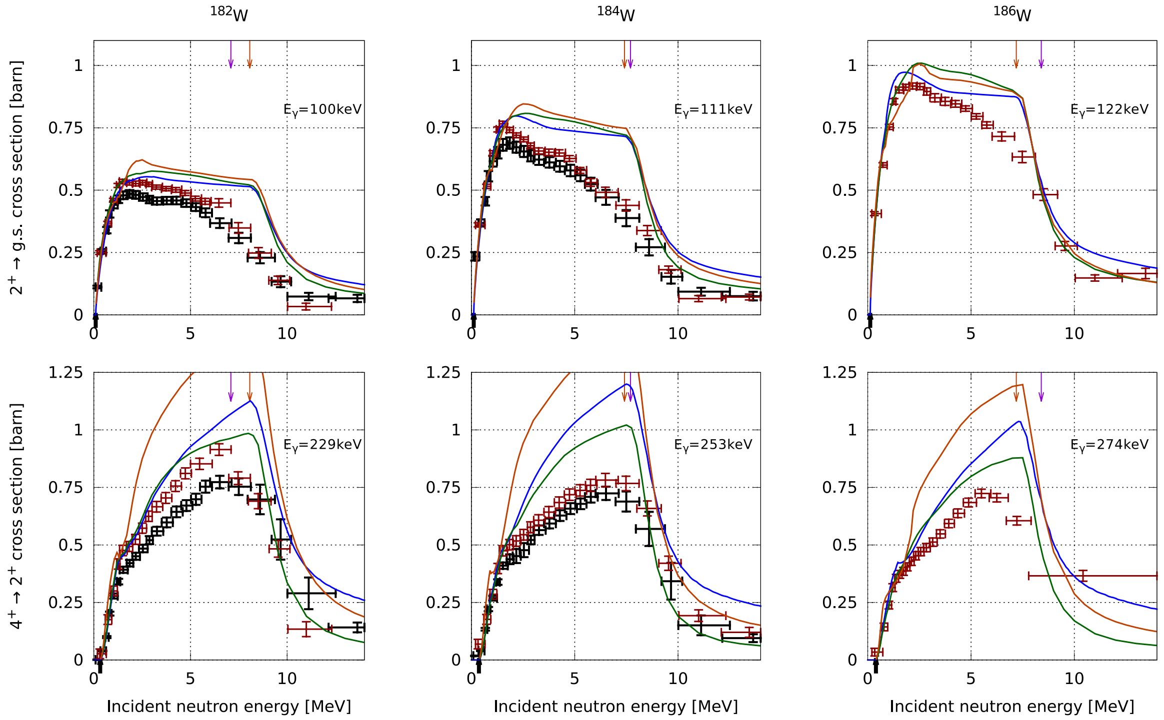

Figure 1. (n, n$'\gamma$) cross sections measured with GRAPhEME for the isotopes ^{182}W (left), ^{184}W (center), ^{186}W (right), for the first excited state to the ground state transition (top) and the second excited state to the first excited state (bottom). The data from isotopic targets are represented with black crosses, the data from the ^{nat}W target in red. It is compared to TALYS-1.73 (full blue line), EMPIRE (dashed orange line) and CoH3 (dotted green line) calculations. For references the energy of the level from which the γ rays are decaying is marked with the black up-pointing arrow on the bottom axis. The neutron separation energy S_n and proton separation energy S_p are marked with the orange and purple down-pointing arrows at the top, respectively.

between the first excited band and the fundamental band [13–15]. In fact, more than 20 transitions are studied for each isotope, decaying from levels as high in excitation energy as 1.5 MeV, within and between several excitation bands. Data from an isotopic ^{186}W target have been recorded but not analyzed yet. In the plots, the cross section error bars represent one standard deviation uncertainty of a Gaussian distribution. The neutron energy error bars represent the range in neutron energy for which the average cross section is given by the point. Some overlap in x-error bars may occur, reflecting the uncertainty in neutron energy selection. For ^{182}W transitions studied in the ^{nat}W target, some (n, 2nγ) reactions on the ^{183}W present in the target lead to a contamination of the ^{182}W(n, n$'\gamma$) cross section above S_n. For the $2^+ \rightarrow 0^+$ and $4^+ \rightarrow 2^+$, a correction was performed by removing the (n, 2nγ) contribution calculated by TALYS-1.7 with default parameters to the experimental data. For the other transitions, the ^{182}W data from the natural target is not shown above 8 MeV. Some amplitude variation between transitions in the different isotopes can be observed, in particular for transitions where $E_\gamma \lesssim 300$ keV, because of internal conversion (IC). This effect is particularly visible for the transitions in the rotational ground state band. Indeed, the moment of inertia decreases with the number of neutrons, leading to higher E_γ for the heavier isotopes which are less converted (The ICC is of the order of 3 for 100 keV transitions in W [16]). Note that for interband transitions, the amplitude variation is not due to IC and reflects the physical processes in the reaction. The agreement of experimental data between natural and isotopic target is good, with a systematically lower cross section in the isotopic targets (≈15% lower), pointing to a possible and yet undetermined bias in the number of atoms in the natural target. The study of ^{182}W(n, 2nγ)^{181}W was attempted. Because of a long lived isomer at low excitation energy in ^{181}W ($\frac{5}{2}^-$ state at 365 keV with a 14.6 μs lifetime), the study of transitions in the (n, 2n) channel is complicated and does not yield any conclusive data. There is also such an isomer in ^{183}W ($\frac{11}{2}^+$ state at 309.5 keV with 5.3 s lifetime) and ^{185}W ($\frac{11}{2}^+$ state at 197.4 keV with 1.7 m lifetime). In consequence, we expect some difficulty extracting valuable (n, 2nγ) cross section data in tungsten isotopes.

4. Comparison to model calculations

The experimental data are compared to calculations by three reaction codes: TALYS, EMPIRE and CoH3.

The TALYS-1.73 calculations [17,18], used an optical potential with coupled channels optimized for deformed nuclei (by default, TALYS considers the nucleus spherical, which is not the case of tungsten isotopes). The nuclear structure considered contained 30 discrete levels. Finally the M1 mode was included in the γ-strength function.

The EMPIRE calculations [19,20] were performed using the model parameters described in Refs. [21] and [22]. The parameters were not optimized to describe the (n, xnγ) transitions and further work is needed to get a more reliable output.

For CoH3 [23], a coupled-channels neutron optical potential was used, with nuclear deformation parameters taken from Finite Range Droplet Model. The code uses a Gilbert-Cameron level density and pre-equilibrium spin

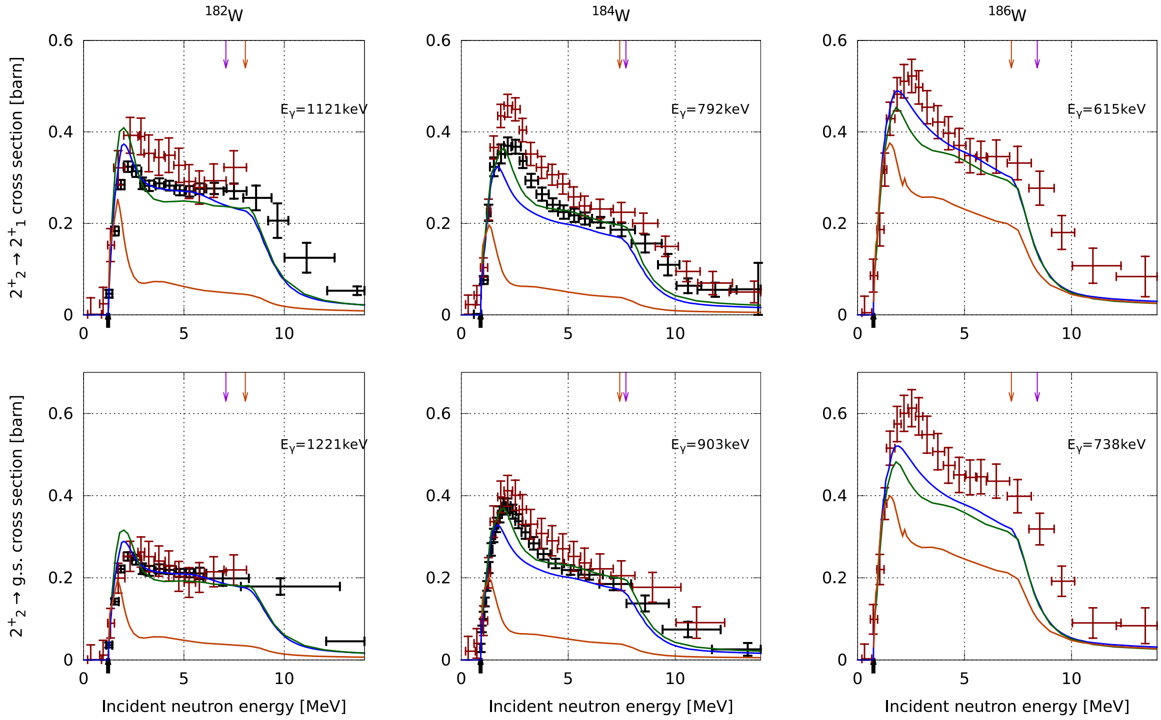

Figure 2. Same as 1 for interband transitions decaying from the 2^+ band. Data for ^{182}W in the natural tungsten target is not represented above 8 MeV because of γ contamination (see text).

distributions where obtained using Feshbach-Kerman-Koonin (FKK) approach. The calculation used 70 discrete levels with levels included inside the continuum. See Refs. [24] and [25] for more details on CoH3 calculations.

For transitions in the fundamental rotational band the agreement of experimental data with model results is limited. The $2^+ \rightarrow 0^+$ cross section presents a plateau in the calculations while experimental data show the cross section falling from ≈ 7 MeV. This might be partly explained by a not yet quantified uncertainty in the HPGe detectors timing due to non-linear response for low energy γ rays. The impact of high energy discrete states in the calculation is also being explored. For the transition from high spin states in the fundamental rotational band, the TALYS and EMPIRE calculations over estimate the transition cross section by a factor of 2 for the $4^+ \rightarrow 2^+$ and up to 6 fold for the highest observed transition ($8^+ \rightarrow 6^+$, not represented in this paper). This is not observed in the CoH3 calculations. We attribute this discrepancy between the data and TALYS, EMPIRE calculations to the effect of the spin distribution in the entrance channel, computed using phenomenological models. Additionally, the use of level densities to describe the high lying states may be worse than the explicit use of discrete levels in the continuum as done by CoH. The same behavior has been observed in ^{238}U [6]. Microscopic calculations have proven a good way to shift this spin distribution to lower values and resolve this difference. See the contribution "*Microscopic description of direct nucleon emission for neutron scattering off actinides*" by M. Dupuis in this conference. For the interband transitions, EMPIRE calculations show a clear disagreement with experimental data in transitions decaying from the 2^+ band. Additional investigations are needed on these discrepancies. The use of better structure information in mandatory coupled-channel calculations, and the coupling of the levels of excited vibrational bands may also improve the description of measured data. Indeed, (n, n′γ) data are extremely sensitive to structure and decay data. Other transitions from other bands are correctly reproduced. For TALYS and CoH3, the interband transitions as well as for other transitions (not presented in this paper), the average agreement between data and calculations is of the order of ≈ 2 to 3 times the uncertainty[1]. This level of agreement allow us to be confident that the structure information (e.g. the branching ratio between γ rays) used by the codes to compute the transitions cross section is correct. To confirm this, we tried to extract the 2^+ level population cross section in ^{184}W from the γ cross sections, which is independent of branching ratios. Although simple in principle, the extraction of the 2^+ level population cross section from γ transition intensity is tricky as more than 20 transitions are feeding the first excited state of ^{184}W, with a highly fractionned intensity. Because of this, only an upper limit to the level population cross section can be extracted. The obtained value is in good agreement with previous data [9,10] and TALYS calculations, but no further conclusions can be drawn.

5. Conclusions and perspectives

(n, n′γ) cross sections have been measured for ^{182,184,186}W in natural and isotopic targets. The results are compared to the calculations by the codes TALYS, EMPIRE and CoH3. Except for an issue in interband transitions in EMPIRE calculations, experimental data agrees (within 2

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[1] This is computed using the root mean square of normalized deviation, i.e., $\sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(\frac{\sigma_i^{\text{experimental}}-\sigma_i^{\text{model}}}{u_i}\right)^2}$, where u_i is the cross section uncertainty for the i-th data point.

to 3 times the one standard deviation) with calculations. Transitions in the fundamental rotational band are still presenting issues. First, the transitions from high spin states are overestimated by TALYS and EMPIRE, while this behavior is not observed with CoH3. This is also seen in the study of ^{238}U [6], and shows the importance of using a microscopic description in the calculations. Also, the transition from the first excited state to the ground state ($2^+ \rightarrow 0^+$) presents a plateau in the calculation that is not seen in experimental data. This could be due to experimental effect not correctly taken into account in the analysis, to the treatment of the continuum in the calculations, or to the use of an incomplete level scheme. The study of (n, 2nγ) transitions was tried, but the extraction of γ ray cross sections was not possible because of long lived isomers in the odd mass isotopes. Looking at level production cross sections would be helpful, but our data allow only the extraction of an upper limit. Many more transitions have been studied for ^{182,184,186}W isotopes. That will provide an extensive set of data to test the models. In particular, the study of ^{183}W will be a great opportunity to study an odd mass isotope. As this study is very hard for actinides [4], that isotope will be ideal to test the predictability of the models for nuclei with an odd number of protons and odd neutron number neutrons (e.g., ^{235}U).

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