

Lawrence Berkeley National Laboratory

Lawrence Berkeley National Laboratory

Title

Gamma Spectrum from Neutron Capture on Tungsten Isotopes

Permalink

<https://escholarship.org/uc/item/26q2x5f0>

Author

Hurst, Aaron

Publication Date

2010-12-15

GAMMA SPECTRUM FROM NEUTRON CAPTURE ON TUNGSTEN ISOTOPES

A. M. HURST^{*1,2}, N. C. SUMMERS¹, B. W. SLEAFORD¹, and R. B. FIRESTONE²

¹Lawrence Livermore National Laboratory, Livermore, California 94551, USA

²Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

*Corresponding author. E-mail : AMHurst@lbl.gov

Received

Accepted for Publication

An evaluation of thermal neutron capture on the stable tungsten isotopes is presented, with preliminary results for the compound systems ^{183,184,185,187}W. The evaluation procedure compares the γ -ray cross-section data collected at the Budapest reactor, with Monte Carlo simulations of γ -ray emission following the thermal neutron-capture process. The statistical-decay code DICEBOX was used for the Monte Carlo simulations. The evaluation yields new γ rays in ¹⁸⁵W and the confirmation of spins in ¹⁸⁷W, raising the number of levels below which the level schemes are considered complete, thus increasing the number of levels that can be used in neutron data libraries.

KEYWORDS : ^{183,184,185,187}W, γ -ray cross sections, neutron capture, EGAF, DICEBOX, level density, photon strength function

1. INTRODUCTION

Improved neutron-capture γ -ray spectra are needed in a variety non-proliferation programs, e.g. in neutron-transport codes for applications in the interest of national security. Building upon known decay schemes and improving the γ -ray spectrum in the neutron data libraries will provide the extensive data essential for modeling and identification of special nuclear materials (actinides) and associated materials (e.g. structural materials). In this paper, we describe improvements to the known decay schemes of the tungsten isotopes from neutron capture onto all but the least abundant of the stable tungsten isotopes, ¹⁸⁰W (natural abundance: 0.12 %). The neutron-capture γ -ray decay data for the compound systems ^{183,184,185,187}W from the *Evaluated Gamma-ray Activation File* (EGAF) [1], has been evaluated by comparing with simulated populations to previously observed levels in these isotopes based on statistical-model calculations using the Monte Carlo code DICEBOX [2]. These evaluations provide improved databases of γ spectra, improved level schemes with clarification of nuclear structure issues such as tentative/unknown spin-parity (J^π) assignments, and will ultimately be used to provide new measurements of the total radiative thermal neutron-capture cross sections (e.g. see Ref. [3]). Furthermore, the results of these evaluations will provide input to the Evaluated Nuclear Data File (ENDF), a nuclear reaction database that is used by the Monte Carlo Neutron Transport (MCNP) codes.

2. THE EGAF PROJECT

This work uses experimental neutron-capture γ -ray cross section σ_γ data from the EGAF database [1] to compare with simulation (see Sec. 3.). Precise values of thermal σ_γ have previously been measured with guided neutron beams (thermal flux $\sim 2 \times 10^6 \text{ s}^{-1} \text{ cm}^{-2}$) at the 10-MW Budapest reactor for all elemental targets with $Z = 1 - 83, 90, \text{ and } 92$, except for He and Pm, in addition to the radioactive targets ⁹⁹Tc and ¹²⁹I [4,5]. Details of a typical experimental setup are described in Ref. [3]. Internal cross sections were calibrated using stoichiometric compounds containing elements with well-known cross sections, e.g. H, N, Cl, and S, in addition to homogeneous mixtures, such as H₂O and HCl, and activation products with well-known γ rays, e.g. ¹⁹F, ²⁸Al, ¹⁰⁰Tc, and ²³⁵U.

3. DICEBOX

The Monte Carlo statistical-decay code, DICEBOX [2], has been used to model the thermal neutron-capture γ cascade by calculating the intensities of transitions feeding the low-lying states. The code generates level-scheme simulations (termed *nuclear realizations*) following the thermal neutron-capture process according to the choice of input statistical-model level density and photon strength functions, and is further constrained by the γ -ray transition probabilities along with known nuclear structure properties of the investigated isotope in the Evaluated Nuclear Structure Data File (ENSDF) database [6]. The DICEBOX

code also uses internal conversion coefficient data from Band Raman (BrIcc) [7]. In the following calculations, A critical energy, E_{crit} , is defined as the energy level, below which, all information in the decay scheme is unique and complete. The code then takes all energy-level and γ -ray branching information beneath E_{crit} from experimental data. The known level scheme is taken from RIPL-3 [8] (Reference Input Parameter Library) with E_{crit} taken as the energy of the level which is the minimum of N_C (the number of levels for which the spins and parities are unique) and N_{max} (the maximum number of levels for which the level scheme is complete). The level scheme is then supplemented with the new information from EGAF. Supplemental feeding to these measured discrete lines from the quasi continuum (i.e. above E_{crit}) is simulated by DICEBOX. All levels and γ rays in the quasi continuum are randomly generated depending on the input level density and photon strength function models selected. In this work 2×10^4 capture state γ -ray decay cascades were randomly generated per nuclear realization using the Monte Carlo method. Measured cross sections of primary transitions (i.e. a transition depopulating the capture state) to levels below E_{crit} were used in each nuclear realization. Good statistical variation in the simulated level feedings was achieved by averaging over five separate nuclear realizations.

Along with the adopted experimental level scheme below E_{crit} and the capture-state spin composition, J , where $J = 1/2^+$ for capture onto even-even targets, and $J = J_{\text{gs}}(\text{target}) \pm 1/2$ for capture onto odd- A and odd-odd targets, the population of the low-lying levels is largely influenced through the choice of level-density and photon strength function models for the different transition types. In the following simulations (Sec. 4.) the back-shifted Fermi gas model [9,10] with von Egidy parameterization [11] was adopted.

DICEBOX only considers photon strength functions for electric dipole ($E1$), magnetic dipole ($M1$), and electric quadrupole ($E2$) transition types. Of these transitions, $E1$ are by far dominant, followed by $M1$, while $E2$ are considerably less significant. A form of the generalized Lorentzian model has been used to describe the shape of the giant electric dipole resonance (GDR) in the present calculations.

The parameterization sets for the $E1$ GDRs for the tungsten isotopes were extracted from experimental data compiled in the RIPL-2 [12] database. For the $M1$ GDR photon strength function, a compound model accounting for scissors-type and spin-flip type excitation modes was adopted, with the resonance-shape parameterization estimated from a previous study of the $^{170,171,172}\text{Yb}$ isotopes [13]. Finally, a giant electric quadrupole photon strength function model describing both isoscalar and isovector vibrational modes was adopted for the $E2$ transitions [14].

4. RESULTS

The results of the calculations simulating the thermal-capture γ cascade using the DICEBOX code are presented

in this section. Modeling of the thermal-neutron capture process onto the four most abundant of the naturally-occurring tungsten isotopes, ^{182}W (26.5 %), ^{183}W (14.3 %), ^{184}W (30.6 %), and ^{186}W (28.4 %), has been used to evaluate the known γ spectrum and make improvements to the decay schemes in the corresponding $^{(A+1)}\text{W}$ compound systems. With the exception of the ^{184}W compound system, all other compounds are formed from neutron capture onto an even-even target, accordingly the capture state is $J^\pi = 1/2^+$ for these systems.

The results of the simulated populations are shown in comparison to the experimental depopulations in the following population-depopulation plots presented in log-log space. Uncertainties along the horizontal axis (experimental depopulation) correspond to experimental errors, while those on the vertical axis (simulated population) are due to Porter-Thomas [15] fluctuations in generating the different nuclear realizations. If the level scheme of the isotope is complete and unique up to the defined E_{crit} , and the decay is purely statistical (i.e. no collective effects), then all data points should fall on the slope of $y = x$. Scatter around this line provides a measure of the quality and completeness of both simulation and experimental data. In the corresponding decay schemes (shown only for ^{185}W and ^{187}W), the width of the arrow illustrates the relative intensity of the transition: the black part of the arrow represents the γ decay and the white part internal conversion; primary transitions are those from the capture state (the highest level, not plotted to scale on the energy axis) represented by blue arrows. New information is highlighted in red on the decay schemes.

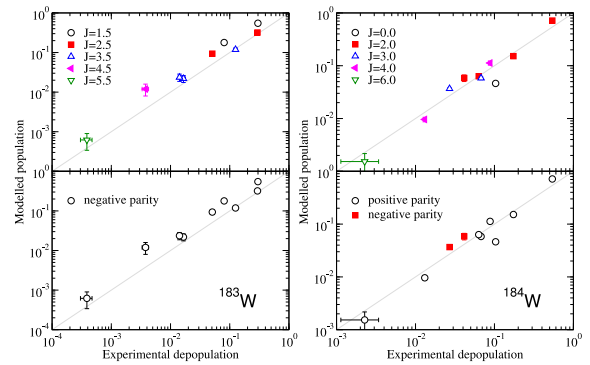


Fig. 1. Simulated population versus experimental depopulation for the compound systems ^{183}W (left) and ^{184}W (right)

4.1 $^{182}\text{W}(n,\gamma)^{183}\text{W}$

Decay data extracted from the RIPL-3 database suggests 10 known levels above the ground state, indicating a value $E_{\text{crit}} = 475.4$ keV for ^{183}W . No further information was available in the EGAF database to allow E_{crit} to be raised. The RIPL-3 and EGAF data compare well beneath E_{crit} . The next highest level at 487 keV with a tentative ($13/2^+$) assignment, is not seen in the EGAF database, due to its high spin. Three primaries have been observed feeding

levels below E_{crit} : a 6190.6-keV $E1$ transition to the $1/2^-$ ground state, a 6144.2-keV $E1$ transition to the $3/2^-$ 46.5-keV level, and a 5981.8-keV $E1$ transition to the $3/2^-$ 208.8-keV level. The calculation shown in Fig. 1(left) uses σ_γ information from the EGAF database; σ_γ data from RIPL-3 does not produce a statistically-significant different result. The deviation of the data from the line is not clear, although, it could point to poor knowledge of the measured γ -ray intensities for some low-energy transitions deexciting low-lying states. This is currently being investigated. Additionally, an unreported $9/2^-$ (308.9 keV) \rightarrow $5/2^-$ (291.7 keV) $E2$ transition has been *implanted* into the level scheme since this would follow a likely rotational sequence. However, the addition/removal of this transition makes no statistical difference to the overall result.

4.2 $^{183}\text{W}(n,\gamma)^{184}\text{W}$

Like ^{183}W , the number of known levels cannot be extended beyond that in RIPL-3. The next level above E_{crit} at 1283.6 keV is tentatively reported as a $(1^-, 2^-)$ state. However, since a corresponding level has not been reported in EGAF it is not possible to extend E_{crit} higher in an attempt to establish the configuration of the 1283.6-keV level. Since the target nucleus, ^{183}W , is odd- A and has a $1/2^-_{\text{gs}}$, the capture state at 7.4117 MeV in ^{184}W can have either a 0^- or 1^- configuration. The present calculation in Fig. 1(right) assumes 100 % capture in the 1^- state. All σ_γ data has been taken from the RIPL-3 file. It should be noted that the RIPL-3 file contains a transition from the 3^- state at 1221.3 keV to the ground state which has not been reported in the EGAF file. Figure 1(right) shows that the only data point deviating significantly from the line is the excited 0^+ state. Various combinations of capture-state fractions are currently being investigated along with σ_γ data from the EGAF database to test the effect on the overall result. The calculation also includes seven primaries feeding states below E_{crit} : 7412.0-keV $E1$ to the 0^+ ground state, 7299.7-keV $E1$ to the 2^+ state at 111.2 keV, 6507.2-keV $E1$ to the 2^+ state at 903.3 keV, 6408.9-keV $E1$ to the 0^+ state at 1002.5 keV, 6289.9-keV $E1$ to the 2^+ state at 1121.4 keV, 6281.5-keV $M1$ to the 2^- state at 1130.0 keV, and 6190.6-keV $M1$ to the 3^- state at 1221.3 keV.

4.3 $^{184}\text{W}(n,\gamma)^{185}\text{W}$

The number of levels below E_{crit} has been increased from 8, in RIPL-3, to 10, based on 2 new levels with corresponding γ -decays in the EGAF data. Figure 2(a) shows the new decay scheme for ^{185}W that has been used to produce the optimal population-depopulation results of Fig. 2(b). The next highest state above E_{crit} , 384 keV ($13/2^+$), due to its high spin, contains no γ -decay information in EGAF. Using DICEBOX, it has been possible to confirm the, previously tentative, $9/2^-$ assignment for the 301.1-keV state, while the postulated tentative ($7/2^-$) assignment from RIPL-3 for the 332.1-keV state has been changed to a $9/2^-$ state on the basis of population-depopulation arguments. Additionally, the energies for both of these states is more accurately measured in EGAF. The RIPL-3 entries for these states were

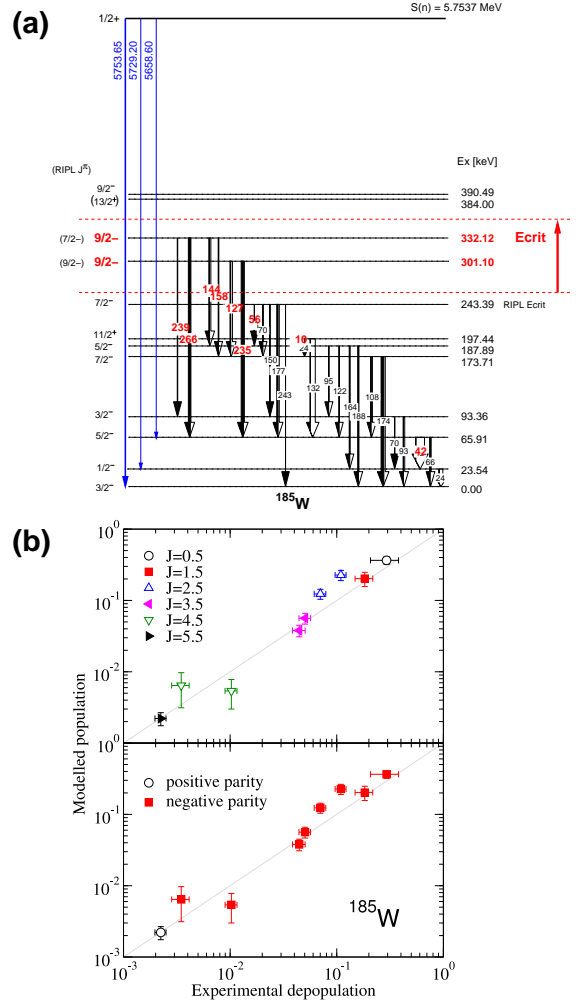


Fig. 2. ^{185}W : (a) decay scheme; (b) simulated population versus experimental depopulation.

reported as 302 keV ($9/2^-$) and 334 keV ($7/2^-$), with no γ -decay information from either state. Four new γ transitions have been reported deexciting the 332.1-keV state and two new transitions deexciting the 301.1-keV state in EGAF. Furthermore, three new γ rays deexciting the lower levels at 243.4 keV, 197.4 keV, and 93.4 keV, have also been reported in EGAF. However, a new transition reported in EGAF from 187.9-keV $5/2^- \rightarrow$ 173.7-keV $7/2^-$ has not been included in the calculation. This very low-energy transition has been reported with a seemingly unrealistic cross section; the largest branch by almost a factor of four. Inclusion of this transition causes the data to deviate significantly from the line in Fig. 2(b). Further investigation of this transition is required to determine the validity of the measured value. EGAF σ_γ data were used for all states in these calculations. It should be noted, however, that for two very low-energy transitions, the reported cross sections were reduced by factors of 10 and 100, respectively, for the 93.4-keV $3/2^- \rightarrow$ 23.5-keV $1/2^-$ and 243.4-keV $7/2^- \rightarrow$ 187.9-keV $5/2^-$ transitions. Again, cross sections for such low-energy transitions are

