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Improving Capture-gamma Libraries for Nonproliferation Applications

B. L. Sleaford, A. M. Hurst

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Correlated Nuclear Data in Fission Events

Venture PI: Patrick Talou, Los Alamos National Laboratory

Improving capture-γ libraries for nonproliferation applications

Brad W. Sleaford (PI)[1] and Aaron M. Hurst[2,3]

[1] *Lawrence Livermore National Laboratory*

[2] *Lawrence Berkeley National Laboratory*

[3] *Department of Nuclear Engineering, University of California, Berkeley*

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Abstract

This report describes the measurement, evaluation and incorporation of new γ-ray spectroscopic data into the Evaluated Nuclear Data File (ENDF) for nonproliferation applications. Analysis and processing techniques are described along with key deliverables that have been met over the course of this project. A total of nine new ENDF libraries have been submitted to the National Nuclear Data Center at the Brookhaven National Laboratory and are now available in the ENDF/B-VIII.beta2 release. Furthermore, this project has led to more than ten peer-reviewed publications and provided theses for five graduate students. This project is a component of the NA-22 venture collaboration on "Correlated Nuclear Data in Fission Events" (LA14-V-CorrData-PD2Jb).

1 Overview

The principal aim of this project is to add new γ-ray spectroscopic data (high-resolution HPGe-quality data) to the Evaluated Nuclear Data File (ENDF) [1] libraries for several high-priority isotopes that will enhance transport-modeling applications. This project leverages heavily upon an existing atlas of data and targeted new capture-γ measurements at reactor facilities that were initiated as an International Atomic Energy Agency (IAEA) Coordinated Research Project (CRP) to develop the Evaluated Gamma-ray Activation File (EGAF) [2].

2 Background and problem

High-resolution observation of γ rays produced in neutron-capture reactions (and inelastic scattering) provide an unambiguous fingerprint of the isotopes within an unknown sample. Nondestructive-assay (NDA) applications may exploit this phenomenon using passive interrogation if spontaneous-fission neutrons are present, or active interrogation where an external neutron source is used to probe the sample. The brightest high-energy γ-ray transitions ($E_\gamma \approx 3 - 12$ MeV) in thermal-neutron capture reactions are those that originate at the capture state, referred to as "primaries." Primary γ rays are often easily seen in spectra from unknown assemblies and clearly indicate the presence of Special Nuclear Materials (SNM; ^{235,238}U and ^{239}Pu), fission products and an array of materials frequently associated with SNM. However, NDA applications are predicated on accurate data and there are well-known data gaps in the neutron-capture γ-ray line data in the ENDF libraries that limit these capabilities. The actinides, for example, have no high-energy capture-γ lines at all, but there are widespread problems elsewhere also.

Throughout this NA-22 project we have taken measures to counter known deficiencies in the ENDF libraries in two distinct regions of the capture-γ spectrum: (i) at high energy where $E_\gamma \gtrsim 3$ MeV; (ii) at low energy where $E_\gamma \lesssim 100$ keV. Our work on tungsten [3, 4, 5] and rhenium [6], in particular, highlights both of these issues. Primary γ rays were identified for the first time in the ^{180}W(n,γ) measurement [4] (Fig. 1), while 50 new primaries were assigned to the ^{186}Re decay scheme via the ^{185}Re(n,γ) measurement [6] (Fig. 2). Because the high-energy regime of the capture-γ spectrum is a region we can delineate and understand completely, as shown in Figs. 1 and 2, these spectra provide enormous potential benefit as an auxiliary forensics tool. Our enriched-sample tungsten and rhenium measurements, both high-Z high-ρ materials, also demonstrated significant γ-ray attenuation that is at odds with much of the existing partial γ-ray production cross-section data [7]. We developed an analytical procedure [5], now tried and tested [6], to correct for this effect (Fig. 3). This problem highlights concerns over much of the existing low-energy capture-γ data for high-density materials that are used to source the ENDF library.

In addition, the white paper (also edited by one of the authors (AH) of this document) that resulted from the *"Nuclear Data Needs for Capabilities and Applications"* workshop [8], held at the Lawrence Berkeley National Laboratory in May, 2015, identified $A \approx 143$ fragments with high yields from thermal, fast, and 14-MeV neutron-induced fission requiring improved spectroscopic data. These data are essential for accurate prediction of inventory in used nuclear fuel assemblies and development of improved physics models for calculation-

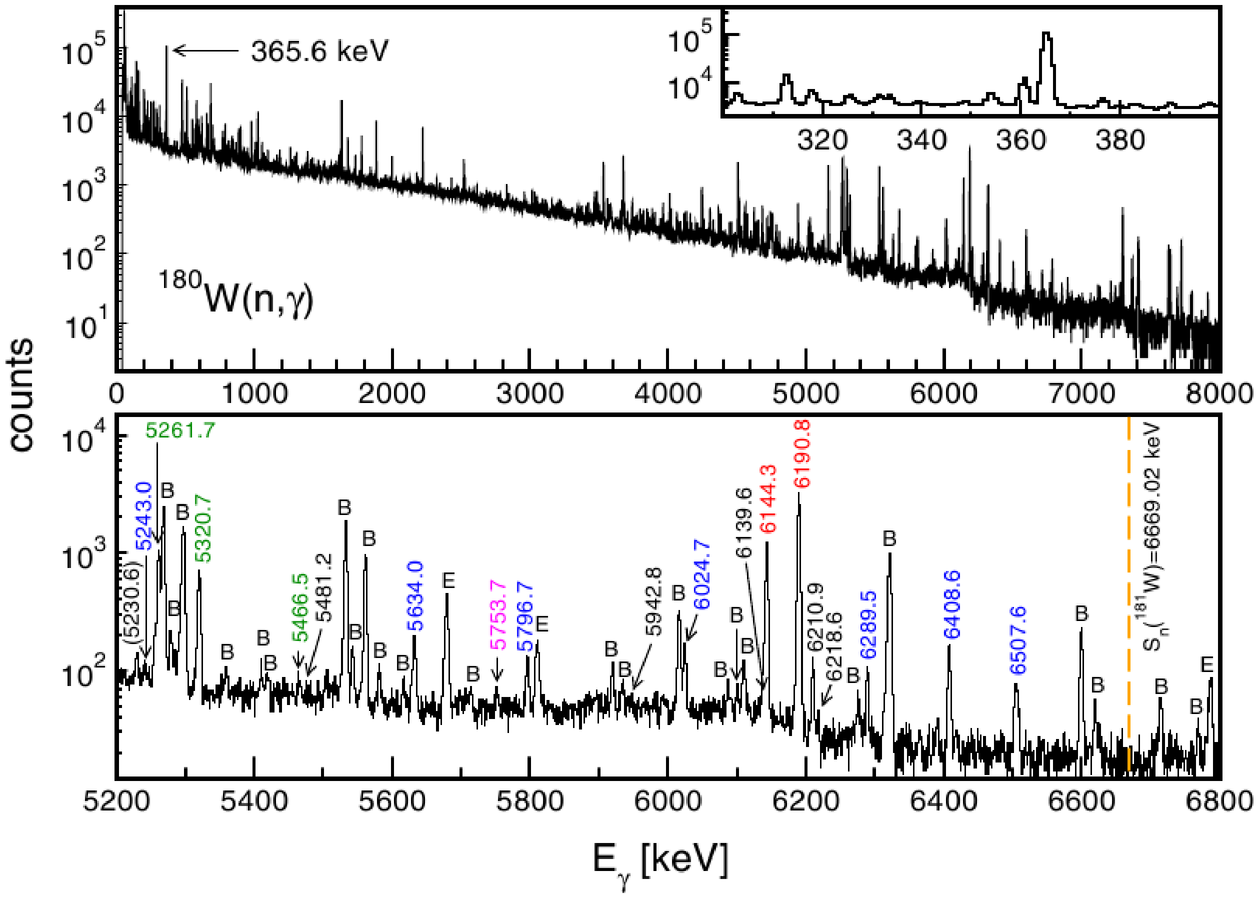

Figure 1: Representative prompt γ-ray spectra from an 11.35% enriched-^{180}W sample showing primaries from ^{180}W(n,γ) for the first time (black labels). For details see Ref. [4].

based nuclear forensics. In light of this application, we have analyzed the ^{139}La$(n,\gamma)^{140}$La prompt spectrum and derived a new set of γ-ray energies and partial cross sections, with improved branching ratios for many of the observed levels, that we are currently preparing for publication [9]. A summary of our other publications related to this topic area is presented in Sect. 4.2.

3 Technical approach

The end goal of this project is to augment the ENDF library with new and improved γ-ray spectroscopic line data. Here, we briefly outline the methodology underpinning this process:

- Partial γ-ray production cross sections (σ_γ) for a particular isotope or element, selected according to a high-priority list, are measured at the Budapest Research Reactor or sourced from EGAF [2].
- These σ_γ data are validated by comparison with theoretical predictions using the sta-

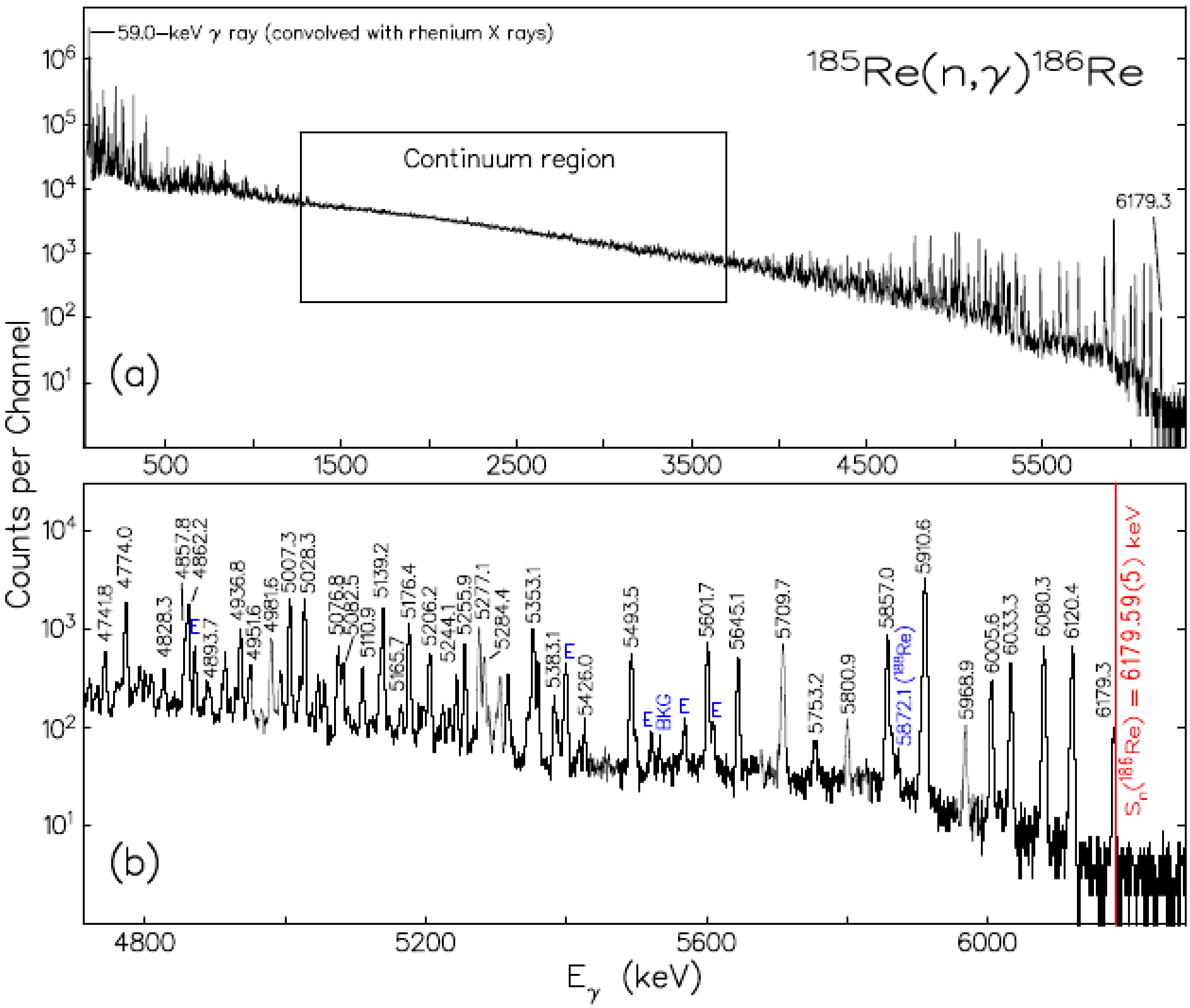

Figure 2: Representative prompt γ-ray spectra from ^{185}Re(n,γ) revealing many new primary γ rays. Taken from Ref. [6].

tistical model for γ decay (DICEBOX [10]) to calculate a system of partial widths for a series of γ cascades:

$$\langle \Gamma_{if}^{XL} \rangle = \frac{f^{(XL)}(E_\gamma) \cdot E_\gamma^{2L+1}}{\rho(E_i, J_i, \pi_i)}. \qquad (1)$$

Here, $\rho(E_i, J_i, \pi_i)$ is the level density at an initial state E_i characterized with a spin-parity $J_i^{\pi_i}$, $f^{(XL)}$ is the photon strength function for a multipole of order L where X denotes electric (E) or magnetic (M) character, and E_γ is the γ-ray energy.

- The validated σ_γ data are then processed into the correct format for incorporation into ENDF and correlations with other sections of the library are verified.

- The LANL and LLNL validation and verification codes (e.g., PREPRO [11], NJOY [12], and FUDGE [13]) are then used to check the integrity of the library.

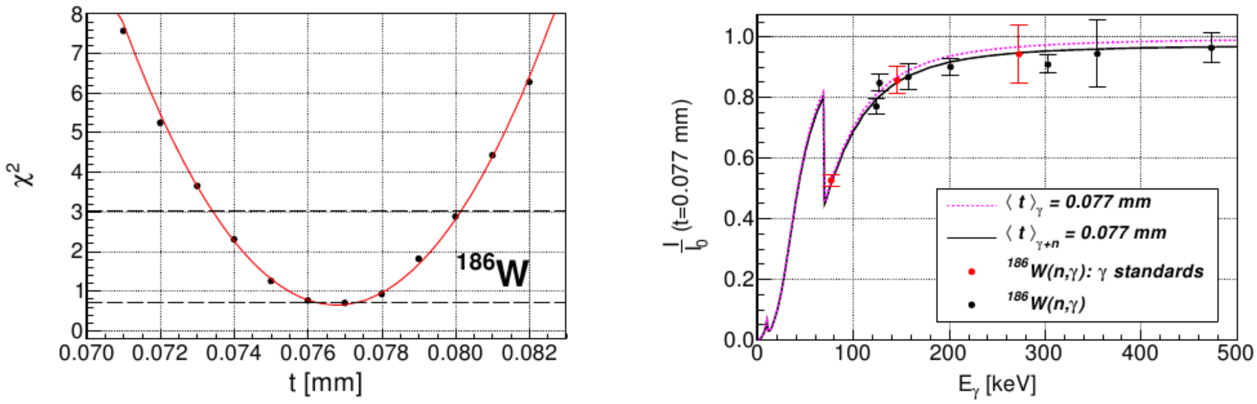

Figure 3: The χ^2-minimization procedure used to determine an analytical correction factor for γ-ray self attenuation in the high-ρ sample ^{186}W. Taken from Ref. [5].

- After generating successful transport-simulation output (MCNP), the libraries are then sent to the National Nuclear Data Center (NNDC) at the Brookhaven National Laboratory (BNL) for further testing and ultimately disseminated in the next ENDF/B-VIII.0 [14] release.

4 Deliverables

4.1 Improving the ENDF libraries

In FY2015 through FY2016 new and improved neutron-capture γ-ray line data were incorporated into and used to update 9 ENDF libraries. These libraries, listed in Table 1 have passed the aforementioned testing requirements in Sect. 3 including generation of representative MCNP output at LLNL, and are now available in the repository for **ENDF/B-VIII.beta2** [14] via NNDC/BNL.

Z	Element	A	Abundance [%]	No. (n,γ) lines
3	Li	6	7.589	3
3	Li	7	92.411	3
5	B	11	80.18	13
9	F	19	100	166
11	Na	23	100	244
13	Al	27	100	290
14	Si	28	92.2297	56
17	Cl	35	75.771	382
17	Cl	37	24.229	76

Table 1: ENDF/B-VIII.beta2 libraries updated with new evaluated capture-γ lines.

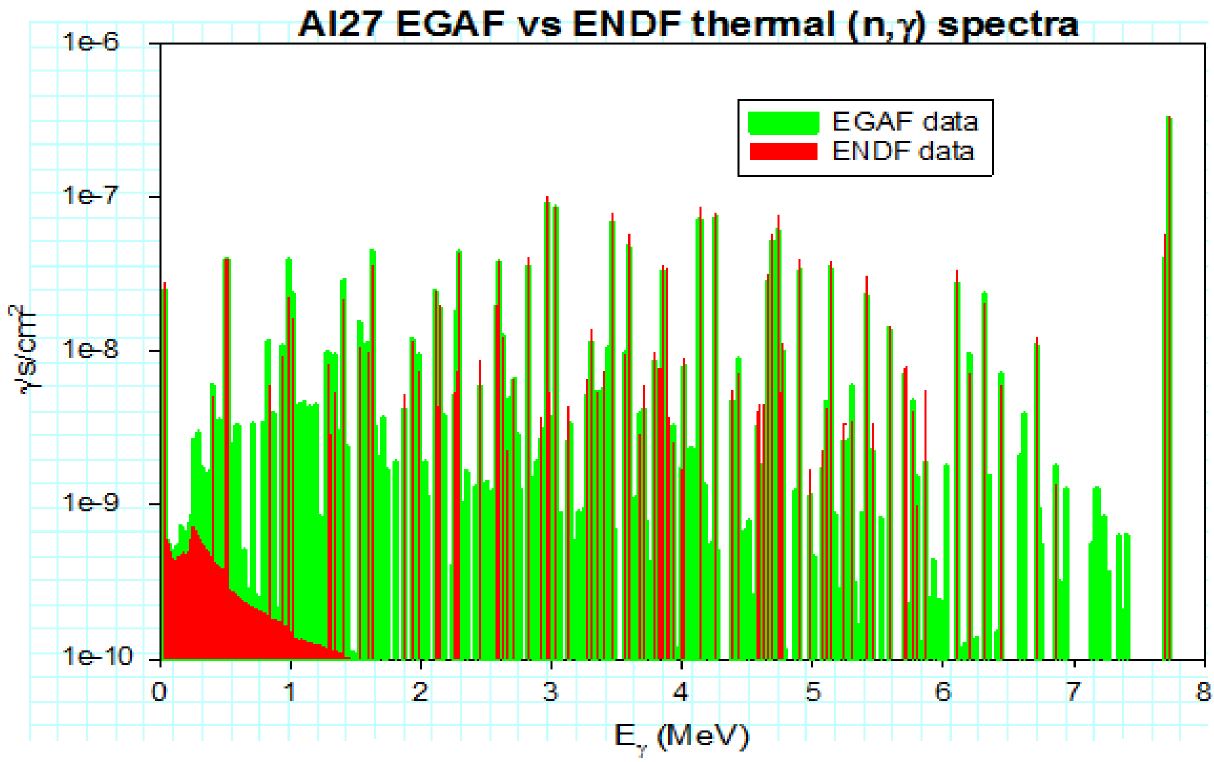

Figure 4: New $\sigma_\gamma(E_\gamma)$ data (green) reveal marked improvement over previous capture-γ data (red) in ENDF for ^{27}Al(n,γ).

High-resolution spectroscopic data are needed for accurate simulations of interrogation systems. As an example, the spectrum in Fig. 4 clearly emphasizes the improvement in quantity and quality attained through incorporation of the new ^{27}Al(n,γ) line data into ENDF over the previous capture-γ data.

4.2 Peer-reviewed publications

This part of the venture is a low-risk project that has produced many peer-reviewed publications. Table 2 summarizes the key projects our collaboration has produced over the past 2 years. In addition to evaluating the partial γ-ray cross sections, our statistical-model analysis permits improvements to the nuclear structure data in decay schemes, e.g., finding new γ rays and J^π assignments. For example, preferential J^π assignments in ^{187}W are illustrated by comparison of modeled and experimental data from ^{186}W(n,γ) shown in Fig. 5; closest proximity to the slope of unit gradient indicate good agreement between model and experiment for an adopted decay scheme. These improvements are also important for many libraries in the nuclear data pipeline because updates communicated to the Evaluated Nuclear Structure Data File (ENSDF) [15] are later reformatted into the Reference Input Parameter Library (RIPL) [16], which in turn is used to source ENDF.

Measurement	Reference	Measurement	Reference
^{139}La(n,γ)	[9]	^{93}Nb(n,γ)	[18]
^{185}Re(n,γ)	[6]	^{23}Na(n,γ)	[19]
Methodology	[5]	^{182,183,184,186}W(n,γ)	[3]
^{180}W(n,γ)	[4]	^{155,157}Gd(n,γ)	[20]
^{242}Pu(n,γ)	[17]	^{152,154}Eu(n,γ)	[21]

Table 2: Selection of peer-reviewed publications from the last two years of this NA-22 project.

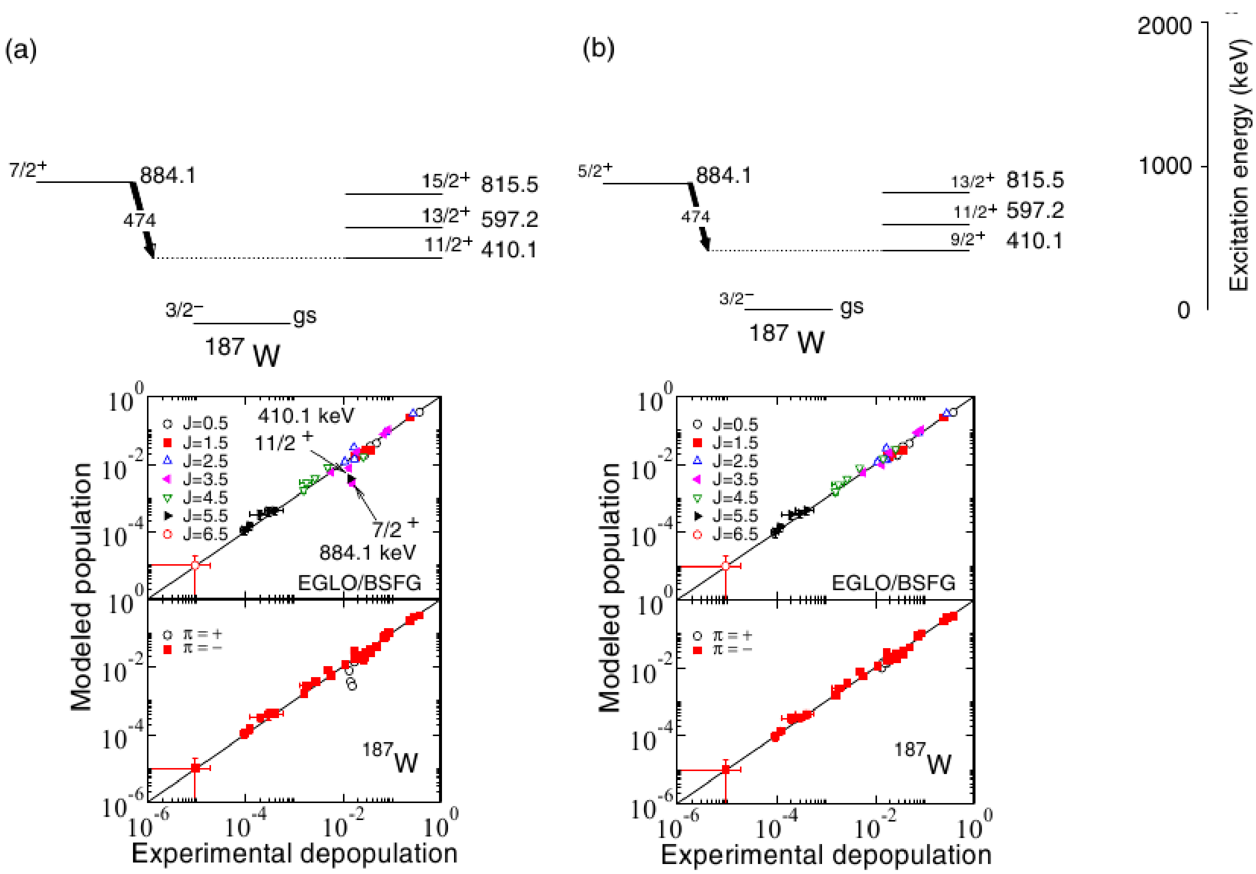

Figure 5: Optimizing the J^{π} assignments in ^{187}W using statistical-model calculations. Taken from Ref. [3].

Student	Degree/Thesis	Institute	Position
Ms. Adriana Ureche	Ph.D. ^{139}La(n,γ)	UC Berkeley [NSSC]	Year 2
MAJ David Matters	Ph.D. (10/2016) ^{185,187}Re(n,γ)	AFIT [DTRA]	Nuclear Effects DTRA (J9NTE)
Danyal Turkoglu	Ph.D. (11/2104) ^{93}Nb(n,γ)	Ohio State University	Postdoc NIST
Christoph Genreith	Ph.D. (10/2014) ^{242}Pu(n,γ)	Jülich	Postdoc FRM-II
MAJ Andrew Lerch	M.S. (02/2014) ^{185}Re(n,γ)	AFIT [DTRA]	Branch Chief Nuclear Survivability DTRA (J9NTS)

Table 3: Graduate students, past and present, that we have mentored on this project.

4.3 Student training

Another area where we have made a significant impact is mentoring and training the next generation of applied nuclear scientists. Over the course of this project we have developed a proven track record of providing graduate students with thesis topics for their research and are actively engaged with students at UC Berkeley through the Nuclear Science and Security Consortium (NSSC) in addition to the Air Force Institute of Technology (AFIT), Ohio State University (OSU), and the Forschungszentrum Jülich in Germany. As summarized in Table 3, our outreach programs and activities have vectored graduates towards careers in applied nuclear science, including the Defense Threat Reduction Agency (DTRA).

Acknowledgments

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