MEASUREMENTS, EVALUATION, AND VALIDATION OF TA-181 RESOLVED AND UNRESOLVED RESONANCE REGIONS

Jesse M. Brown

Submitted in Partial Fulfillment of the Requirements for the Degree of

DOCTOR OF PHILOSOPHY

Approved by:
Yaron Danon, Chair
Emily Liu
Wei Ji
Vladimir Sobes
Hyun Gook Kang
Devin Barry

Department of Mechanical, Aerospace, and Nuclear Engineering
Rensselaer Polytechnic Institute
Troy, New York

[Aug 2019]
Submitted June 2019
# CONTENTS

LIST OF TABLES ................................................................. vi

LIST OF FIGURES ............................................................... viii

ACKNOWLEDGMENT .............................................................. xvii

ABSTRACT ........................................................................... xix

1. INTRODUCTION ............................................................... 1
   1.1 Nuclear Data ............................................................... 1
   1.2 Time-of-Flight Measurements ....................................... 4
      1.2.1 Neutron Transmission .......................................... 5
      1.2.2 Neutron Radiative Capture ................................... 7
   1.3 Nuclear Data Evaluation .............................................. 8
   1.4 Tantalum: Background and Motivation ........................... 9
   1.5 Validation Transmission ............................................ 15

2. NEUTRON-NUCLEUS INTERACTIONS .................................. 19
   2.1 RRR ........................................................................... 19
   2.2 Resonance Self-Shielding ............................................ 24
   2.3 URR .......................................................................... 27
   2.4 Programming for the URR .......................................... 30

3. MEASUREMENTS .............................................................. 32
   3.1 Evaluation Data .......................................................... 33
      3.1.1 100 m Transmission ............................................. 33
         3.1.1.1 Beam Characteristics ...................................... 34
         3.1.1.2 Neutron Transmission Detector ....................... 34
      3.1.2 45 m Capture ..................................................... 36
         3.1.2.1 Beam Characteristics ...................................... 37
         3.1.2.2 Neutron Capture Yield Detector ....................... 38
   3.2 Validation Data: Thick Sample Transmission ................. 41
      3.2.1 Beam Characteristics ........................................... 43
      3.2.2 Neutron Transmission Detector ............................ 44
4. DATA REDUCTION ......................................................... 46
  4.1 Neutron Transmission ............................................. 46
     4.1.1 Dead Time .................................................. 47
     4.1.2 Background ................................................ 48
        4.1.2.1 Background: 100 m Transmission .................... 50
        4.1.2.2 Background: 35 m Thick-Sample Transmission .... 51
     4.1.3 Uncertainty and Correlation ............................. 53
     4.1.4 Reporting Data Covariance ............................... 56
  4.2 Neutron Radiative Capture ....................................... 58
     4.2.1 Background ................................................ 59
     4.2.2 Weighted Neutron Capture Yield ......................... 59
     4.2.3 Normalization of Capture Yield ......................... 62
     4.2.4 Uncertainty and Correlation ............................. 65
     4.2.5 Photon Attenuation ...................................... 66
        4.2.5.1 Modeling the Photon Attenuation Correction ..... 67
        4.2.5.2 Fitting the $C_1$ Parameter ....................... 69
5. RESULTS & EVALUATION ............................................... 74
  5.1 RRR .............................................................. 74
     5.1.1 SAMMY Program .......................................... 74
     5.1.2 RRR Results .............................................. 75
     5.1.3 Resonance Statistics .................................... 82
     5.1.4 RRR Data Format ........................................ 85
  5.2 URR .............................................................. 86
  5.3 Validation Transmission ......................................... 93
     5.3.1 MCNP ...................................................... 93
     5.3.2 Results ................................................... 93
     5.3.3 Improving the Model .................................... 97
        5.3.3.1 Modification of Existing Parameters ............. 98
        5.3.3.2 Extension of the RRR ............................... 100
        5.3.3.3 Multiple Fits of the URR ......................... 102
     5.3.4 Effect on Criticality ................................... 106
6. CONCLUSIONS .......................................................... 112
  6.1 $^{181}$Ta Evaluation ........................................... 112
  6.2 Differential Validation Transmission .......................... 113
6.3 Future Developments ....................................................... 114
  6.3.1 Thermal Data .......................................................... 114
  6.3.2 Upgrade of the Capture Detector ................................. 115
    6.3.2.1 Angular Distributions .......................................... 115
    6.3.2.2 Detector Mounting System ................................. 116
    6.3.2.3 Background Shielding .................................... 117
  6.3.3 Upgrade of the Transmission Detector ......................... 117

BIBLIOGRAPHY .............................................................. 119

APPENDICES

A. Programming & Input Files ............................................. 129
  A.1 Fortran Program: Sampling the Wigner Distribution ............ 129
  A.2 Transmission Benchmark MCNP Input File ......................... 130
  A.3 FITACS Input File .................................................. 135
  A.4 SESH Input File .................................................... 138

B. Sample Characteristics ............................................... 140

C. Useful Derivations .................................................... 142
  C.1 $\frac{\partial E}{\partial t}$ for Relativistic Kinetic Energy ........ 142
  C.2 $\frac{\partial t}{\partial E}$ for Relativistic Kinetic Energy ........ 143

D. Cross Section Reconstruction with NJOY21 ....................... 145

E. Covariance Matrix Input .............................................. 147

F. Determining Uncertainty on Monitor Counts ......................... 151

G. Copyright Permissions ................................................. 153
LIST OF TABLES

1.1 The boundaries for the end of the RRR and URR for each of the libraries discussed during this work. .......................................................... 9

3.1 Summary of measurements performed. The discussion of the details listed here is relegated to the proceeding sections of this chapter. .......................... 32

3.2 The voltages, lower level discriminator (LLD), and upper level discriminator (ULD) settings for the MELINDA system. The signal of the PMTs for each module is summed and passed through the same discriminator logic. ........ 34

3.3 The voltages for the PMTs in the 45 m capture detection system during the Aug 2017 Ta measurement. ................................................................. 39

3.4 The voltages for the PMTs in the 35 m transmission detector system, along with the associated discriminator settings for the system. The signal from both PMTs is summed and passed through the same discriminator logic. ........... 45

5.1 The measurements used in this work to fit new resonance parameters using the evaluation tool SAMMY 8.1. .................................................. 76

5.2 Various average level parameters used to describe the URR from 2-120 keV. The RPI parameters taken from the resonance parameters fit with SAMMY are listed for those which could be measured. The initial parameters used for the URR fit in FITACS (marked by “I”) take the RRR average parameters where possible; see the text for discussion of the other parameters. The final fitted parameters in the URR (marked by “F”) are the final output from the FITACS fit. The average parameters from McDermott [33] are listed, as well as the average parameters found in the Atlas of Neutron Resonances [55] by Mughabghab. McDermott used the average level density of Mughabghab, as indicated by the *. Subscripts in the \( \langle \text{Par} \rangle \) column indicate quantum angular momentum for \( l = 0, 1, 2 \). Uncertainty is omitted for quantities related to \( a_c \), as \( a_c \) does not have a defined uncertainty. .................................................. 92

5.3 The capture resonance integrals \( (I_c) \) for the various evaluations as calculated by NJOY21 [62]. The ENDF/B-VIII.0 value is greatest, indicating that it will have the greatest neutron absorption. Also note the proximity of the RPI and ENDF/B-VIII.0 values as compared to that of JEFF-3.3. This is consistent with the TEX benchmark results presented in Fig. 5.24. ......................................... 110

B.1 The sample impurities for all the samples obtained in an order from Stanford Advanced Materials, LOT NUMBER: WT170623-5443. .......................... 140
B.2 The sample dimensions and mass are listed for each of the ordered samples. In an effort to be explicit the calculated quantities of density and areal density of the samples are listed as well. ................................. 141

E.1 The input covariance matrix $C_{sy}$ for the systematic uncertainty variables found in the transmission equation (Eq. 4.1). ................................. 147

E.2 The transmission, uncertainty on the transmission, and statistical uncertainty on the sample ($\dot{C}_{Ta,i}$) and open ($\dot{C}_{o,i}$) count rates. ................................. 148

E.3 The first set of derivatives of the transmission equation given in Ch. 4 for the validation transmission. The derivatives occur at the energies listed in Table E.2. 149

E.4 The second set of derivatives of the transmission equation given in Ch. 4 for the validation transmission. The derivatives occur at the energies listed in Table E.2. 150
LIST OF FIGURES

1.1 A diagram to depict how cross section is defined by beam intensity $I$, atomic density $N$, and sample thickness $dx$. The arrows represent the direction orthogonal to the face of the target sample. ............................................... 2

1.2 The total cross section for $^{181}$Ta as evaluated in JEFF-3.3 [16]. RRR represents the resolved resonance region, URR the unresolved resonance region, and Continuum represents the continuum region where the cross section changes slowly as a function of energy. ......................................................... 3

1.3 The percent error in the non-relativistic approximation for kinetic energy as a function of energy. ................................................................. 5

1.4 A diagram of a typical TOF transmission experiment. Electron pulses are accelerated toward a neutron-producing target. The pulsed neutron emission is collimated towards a sample, collimated after the sample, and recorded by a neutron detector at a fixed flight path length. .............................. 6

1.5 A diagram of a typical TOF radiative capture yield experiment. The pulsed neutron beam is collimated towards a sample, which may capture a neutron and emit a cascade of capture photons. Capture photons emitted in the direction of the photon detector are recorded as a function of TOF. .......... 8

1.6 Shown above are the evaluated total cross sections from the JEFF-3.3, ENDF/B-VIII.0, and JENDL-4.0 libraries. The energy regions (RRR: 1-2400 eV, URR: 2.4-100 keV, Continuum: 0.1-10 MeV) are labeled by the model which is used to represent them in JEFF-3.3, and JENDL-4.0. It can be seen that ENDF/B-VIII.0 deviates significantly from the other evaluations: ENDF/B-VIII.0 sets the end of the RRR at 330 eV, and ends the URR model at 5 keV. The JEFF-3.3 and JENDL-4.0 libraries are nearly identical. ........................................ 10

1.7 The normalized computed neutron multiplication factor $C/C_{ENDF}$ for ENDF/B-VIII.0, JEFF-3.3 and JENDL-4.0 cross section evaluations of Ta employed in the preliminary TEX benchmarks [25]. For each criticality calculation, only the cross section library for $^{181}$Ta is changed. This shows the impact of the different evaluated $^{181}$Ta cross sections on epithermal criticality applications. ........... 11

1.8 Energy regions covered by referenced works listed by evaluators in the information file portion of each ENDF file for $^{181}$Ta, and the corresponding data type. The dashed vertical lines delineate the beginning and end of the shared JEFF-3.3/JENDL-4.0 RRR. ......................................................... 12

1.9 A representative selection of available total and capture cross section experimental data for $^{181}$Ta from the IAEA EXFOR [34] database. Data sources are from Refs. [31],[33],[35],[30],[36],[37],[38],[39],[40],[41]. ........................................ 13
1.10 The transmission datasets for $^{181}$Ta made by Harvey [34], found in EXFOR. There are two thicknesses, the thicker of the two shows excellent resolution into the keV region. The thickness of each sample is listed in atoms per barn [at/barn].

1.11 Neutron capture rate as a function of TOF was measured for $^{181}$Ta in the RPI LSDS. The LSDS enables statistically accurate neutron capture measurements with poor energy resolution. Discrepancies between libraries and experimental data can be seen in the URR. The plot on the right shows the spectrum over all energies, the plot on the left shows the energies where the RRR transitions to the URR. Used with permission from Ref. [42].

1.12 Experimental design of the 1972 Fe broomstick transmission by Maerker [48]. Neutrons are emitted from a reactor, collimated towards a cylindrical sample of Fe, and finally collimated further towards a proton recoil detector. The energy of the transmitted neutrons is calculated by the energy deposited from a proton recoil, which has lower energy accuracy than the TOF method. (Note: Not to scale.)

1.13 The approximate geometry from the 2001 publication by Murata on high energy transmission and angular benchmarks using TOF techniques [47]. The geometry is highly collimated with a short flight path of 8 m. (Note: Not to scale.)

1.14 The geometry for the RPI thick sample transmission measurement. This geometry is well collimated and uses a polyethylene neutron reflector to reduce interfering in-beam background. The $^4$B$^4$C and Pb collimator combination contained in the dashed box are repeated four times in succession in the flight path; a detailed CAD drawing for the $^4$B$^4$C and Pb collimation can be found in Ref. [50]. The simple transmission experiment, as applied to URR measurements, can be used by evaluators to validate the resonance self-shielding correction made in transport models of the URR. (Note: Not to scale.)

2.1 Diagram of the total, capture, and scattering cross section as computed by the Breit Wigner formula for the first resonance of $^{181}$Ta at energy $E_\lambda = 4.28$ eV.

2.2 The pointwise transmission based on JEFF-3.3 cross section of $^{181}$Ta and $n=0.01$ is plotted with a solid green line. The average transmission taken over the TOF range 500→2500 $\mu$s results in an $\langle T(\sigma_t) \rangle \approx 0.6$, which was plotted with the upper horizontal dashed green line. Taking the average of the cross section over the same energies, and calculating the transmission of the average cross section results in $T(\langle \sigma_t \rangle) \approx 0.2$, which is given by the lower red horizontal dashed line. This illustrates the importance of accounting for cross section fluctuation in measurements.

2.3 Transmission as a function of the total cross section $\sigma_t$ is plotted for two different sample thicknesses. The greater the sample thickness, the more non-linear the relationship between transmission and $\sigma_t$. The vertical dashed line represents the average cross section $\langle \sigma_t \rangle$. 

ix
3.1 The gated and un-gated MCA spectra are shown with the red and blue lines, respectively. The gated counts come from the voltage pulses in the PMTs which pass the LLD and ULD. The MCA was collected using a 1024 bin structure.

3.2 The Mid Energy LI-glass Neutron Detector Array (MELINDA). Each of the four aluminum cases hold $^6$Li doped scintillating glass, with 2 PMTs coupled to the light-tight aluminum module. The detector resides in the RPI Linac 100 m detector station.

3.3 An illustration of the new reference source position for the 100 m transmission detector MELINDA. This geometry was used to perform a reference count rate for the MELINDA detector before each measurement. Starting from the right: the yellow rectangle represents the 10.16 cm (4 in) polyethylene block with a bore hole that houses the source, followed by a gray block that represents two 5.08 cm (2 in) Pb bricks.

3.4 An image of the new reference source geometry for the 100 m transmission detector MELINDA. The source is kept on the end of a 3.5 m Al rod, which is seen in the images to the left and right. The source position in the shielded geometry is marked by a red circle. This new reference geometry reduced the dose to a researcher by a factor of approximately 2-3.

3.5 The capture detector system at a flight path of approximately 45 m. Four $C_6D_6$ detectors are suspended by aluminum rods and face the sample of interest. Sample material was changed by an automated step motor, which moved the sample holders to the left and right (orthogonal to the neutron beam).

3.6 The measured pulse integral for each discrete Compton edge energy is plotted along with fitted lines for each detector. The energy of the decay gammas are given in parentheses. The parameters from the fit are used in post-processing to determine the energy deposited in the detectors.

3.7 The 35 m $^6$Li doped glass scintillator detection system. The detector resides along the East beam line at the RPI Linac, as indicated by the label on the bottom window sill.

3.8 Two 6 mm thick Ta samples are stacked to form the 12 mm thick sample used in the thick-sample validation transmission measurement.

3.9 The reference source geometry for the 35 m transmission detector. This reference count rate is measured to ensure that the detector is behaving normally before collecting the TOF data of interest. The direct distance from the source to the center of the glass scintillator is approximately 2.6 m. (Note: not to scale.)
4.1 An illustration of the dead time correction employed for the count rates used for the transmission and capture yield measurements. The dead time is indicated by the red object occurring over several TOF bins. Each bin at time $i$ is corrected by $j$ many bins that the dead time spans preceding the midpoint of bin $i$. Dead time is determined for each system, and applied in the same fashion for each.

4.2 Count rates measured by the 35 m transmission detection system. The TOF range shown includes the Co resonance at 132 eV, which removed almost all on-energy neutrons near that energy. This is an example of a notch which was used to measure background in the TOF experiment. This count rate was measured during a dedicated notch run. The discrete background point shown by the red circular marker was calculated from the mean of the count rate in the delineated region.

4.3 The count rates measured by the MELINDA detector at a flight path of 100 m. The background counts and fitted background functions for the “open” and “sample in” configurations are given by the blue and red circles and lines, respectively. The background functions are normalized to the fixed notch (132 eV resonance of Co) at an approximate TOF of 625 µs.

4.4 The count rate for the thick-sample transmission measurement. The upper solid green curve represents the measured open beam count rate and the dashed green curve matching the green circular points is the calculated open background count rate. The solid blue line represents the sample count rate and the dashed blue line matching the blue circular points is the calculated sample background count rate. Blue and green circles indicate “black resonance” count rates for the sample in and open count rates, respectively.

4.5 An example file format for reporting experimental data covariance. The orange box indicates systematic uncertainties ($\Delta k_{Ta}, \Delta k_o$, etc.). The blue box indicates correlated input: a covariance matrix for some of the systematic variables ($\Delta a^2, \Delta a\Delta b$, etc.). The red box contains vectors in the order of: energy, observable ($T$), and uncertainty on observable ($\Delta T$). The green box indicates columns of statistical uncertainty as a function of energy, in this case $\Delta \hat{C}_{Ta,i}$ and $\Delta \hat{C}_{o,i}$. Finally the yellow box indicates columns of derivatives of the transmission with respect to each variable as a function of energy ($\frac{\partial T_i}{\partial k_o}$, $\frac{\partial T_i}{\partial \hat{C}_{o,i}}$, etc). Commented out by a # just above each section of data are descriptive strings.
4.6 The experimental transmission measured at 100 m for a 6 mm thick Ta sample along with the fitted theoretical curve produced by SAMMY 8.1. The fitted parameters from these two resonances are used to normalize the capture measurement at the 45 m capture detector. The deviation of the theoretical curve from the data at the peak of the 208 eV resonance is due to the local variation of the sample background away from the smooth theoretical sample background function (Eq. 4.7). Assuming that the theoretical transmission is correct, and modifying the sample background $k_{Ta} \dot{B}$ for the experimental transmission to match the theory results in an increase on the normalization of the yield data of approximately 2.8%.

4.7 The experimental capture yield measured at 45 m for a 2 mm thick Ta sample along with the initial and fitted theoretical curves produced by SAMMY 8.1. The initial theoretical curve is from the parameters found by fitting the measured transmission of the 1, 3, and 6 mm Ta samples at 100 m. The fitted theoretical curve in this figure comes from allowing the normalization of the yield and the energy of the capture yield resonances to vary in the SAMMY 8.1 fit.

4.8 Illustration of a neutron that was collimated toward a sample, captured, and subsequently emitted a photon. Each exponential neutron flux profile across the sample thickness corresponds to the cross section found on the profile of the resonance. At the peak of a resonance in energy space, the neutron is more likely to capture at the sample surface and the emitted photon more likely to escape the sample and interact with the gamma ray detector. On the wings of a resonance a neutron is more likely to penetrate further into the sample, and the emitted photon less likely to escape.

4.9 The energy spectrum of cascade photons occurring during the de-excitation of a compound $^{182}$Ta nucleus. This spectrum (blue) was generated using the Monte Carlo code DICEBOX [81] by Becvar which uses the RIPL and EGAF databases as input. The DICEBOX spectrum is compared with the ENSDF values (green) published by the NNDC. The red line at 0.150 MeV indicates the lower level discriminator (LLD) of the experimental analysis. The fraction of events above the LLD is $\approx 76\%$.

4.10 The discrete values $k_{a,\text{sim}}(\sigma_{t,i})$ for correction factor $k_a$ as determined by Monte Carlo calculation using MCNP are plotted with Eq. 4.34 fitted to those values.

4.11 The experimental data measured by McDermott [50] for the first 3 resonances in $^{181}$Ta. Plotted in solid lines are the theoretical curves as calculated by SAMMY 8.1 using JEFF-3.3 resonance parameters as input (blue line), and the same theoretical curve multiplied by the correction $k_a$ (red line).

4.12 The experimental data measured by McDermott [50] for the first 3 resonances in $^{181}$Ta. Plotted in solid lines are the fitted theoretical curves as calculated by SAMMY 8.1 without the correction factor $k_a$ (blue line), and the fitted theoretical curve as calculated by SAMMY with the correction $k_a$ (red line).
5.1 Low energy experimental data. The transmission measurements come from the Harvey datasets [34], and the capture yield measurements were made at RPI [33]. The “thin” and “thick” Harvey samples were 0.000167 and 0.002881 at/b, respectively. The lowest energy datum for the “thick” Harvey measurement was $\approx 4.4$ eV in the middle of a resonance, and was fit down to that energy. The legend labels end in “C” or “T” to indicate capture yield and transmission respectively. It should be noted that the 4 eV resonance has the greatest impact on integral quantities for $^{181}Ta$.

5.2 Low energy resonance fitted to experimental data. The transmission measurement comes from the Harvey dataset [34], and the capture yield measurements were made at RPI [33]. The “thin” Harvey sample was 0.000167 at/b in areal density. It should be noted that the 4 eV resonance has the greatest impact on integral quantities for $^{181}Ta$. The agreement of the RPI and Harvey datasets indicates that the $k_a$ correction improved the model of the data.

5.3 Resonances are shown past the ENDF/B-VIII.0 RRR, and are seen to be well resolved. The legend labels end in “C” or “T” to indicate capture yield and transmission respectively.

5.4 The 304 eV resonance is highlighted here by inaccurate resonance parameters published in the JEFF-3.3 evaluation. This discrepancy can be seen in the transmission and capture yield datasets. The RPI fit corrects this inaccuracy to better match the data.

5.5 Near the end of the RRR of JEFF-3.3 the experimental data are very likely to be missing levels, but the Reich-Moore R-matrix model represents the data very well. The legend labels end in “C” or “T” to indicate capture yield and transmission respectively.

5.6 The end of the fitted RRR is shown in greater detail. The 2 mm capture data and 6 mm transmission data are shown in comparison to the yield and transmission as predicted by JEFF-3.3, respectively. The JEFF-3.3 evaluation did not include the 2460 eV resonance. This resonance was fit as part of the RPI evaluation.

5.7 For the both the level spacing and level width plots the plotted distributions are the number of resonances greater than $x_1 = S/D$ and $x_2 = \Gamma_n^0/\langle \Gamma_n^0 \rangle$, respectively. Note that the abscissa of the plots are $\sqrt{x_1}$ and $\sqrt{x_2}$. The top left and right plots show the normalized Wigner distributions for the $J = 3.0$ and $J = 4.0$ level spacing, respectively. The variable $x_1 = S/D$. The Wigner distribution derived from the RPI measurement for $J = 3.0$ had 1 level greater than $\sqrt{x_1} = 2.5$. The bottom left and right plots show the normalized Porter Thomas distributions for the $J = 3.0$ and $J = 4.0$ reduced neutron widths, respectively. The Porter Thomas distribution calculated from the RPI measurement for $J = 4.0$ contains 3 levels greater than $\sqrt{x_2} = 3.5$. 
5.8 The cumulative levels as a function of energy compared to the expected cumulative levels as a function of energy. The slope of the expected cumulative levels is given by $1/D$. $D$ was calculated from the low energy region of 0-300 eV as its behavior diverged from linearity shortly after 300 eV. This non-linearity could be due to missing levels. ................................................................. 85

5.9 The measured average total cross section is shown corrected (black) and uncorrected (blue) for resonance self-shielding by SESH. The JEFF-3.3 evaluated cross section is shown as a reference for the expected cross section prior to this measurement. ................................................................. 88

5.10 A diagram of the iterative process for fitting grouped total and capture cross section in the URR using the SESH program and FITACS program in SAMMY. The data are corrected by evaluated parameters to start, and then the fitted parameters correct the data to be re-fitted until a convergence has been found for the correction factor calculated by SESH. Usually 2-3 iterations achieve convergence. ................................................................. 89

5.11 The measured average capture cross section is corrected for multiple scattering and self-shielding by SESH and fitted using SAMMY/FITACS. The uncertainty on the measured data comes from statistical uncertainty in the count rate and systematic uncertainty in the flux normalization and background subtraction. It was assumed that the flux, background and measured capture rates were not correlated. The dashed curve represents a theoretical cross section using the average parameters calculated from the RRR measurement. ................................................................. 90

5.12 The measured average total cross section is corrected for self-shielding by SESH and fitted using SAMMY/FITACS. The dashed curve represents a theoretical cross section using the average parameters calculated from the RRR measurement. It should be noted that the uncertainty in the measured total cross section includes statistical uncertainty from the count rate and systematic uncertainty deriving from the background subtraction method described in §4.1.2. ................................................................. 91

5.13 The grouped thick-sample transmission measurement in red, along with the MCNP calculated transmission using various evaluated cross section libraries. ................................................................. 94

5.14 The correlation matrix for the $\langle T_iT_j \rangle$ covariance matrix for the thick-sample transmission measurement. Plotted above the correlation matrix is the fractional uncertainty in the transmission measurement, and to the right of the correlation matrix is the corresponding transmission. The two dashed peaks in the fractional uncertainty (up to $\approx 5\%$) are caused by Al resonances at 35 and 88 keV. ................................................................. 96

5.15 A description of the work flow for evaluating measured data and testing how well the new evaluation predicts neutron transmission. The measured data are fitted for resonance parameters, formatted into ENDF/B format files, processed by the NJOY21 program to an ACE file, and finally used by MCNP in a Monte Carlo calculation. ................................................................. 97
5.16 The grouped thick-sample transmission measurement in red, along with the MCNP calculated transmission using various evaluated cross sections. The JEFF-3.3 evaluation was modified to include URR average resonance parameters as reported by McDermott et. al [33], and a new transmission curve using the modified JEFF-3.3 file was plotted in black.

5.17 The grouped thick-sample transmission measurement in red, along with the MCNP calculated transmission using various evaluated cross sections. The JEFF-3.3 evaluation was modified to include RRR and URR parameters from the analysis described in §5.1 and §5.2, respectively. The new transmission curve using the modified JEFF-3.3 file was plotted in black.

5.18 The measured transmission and capture yield data from the RPI measurement. The fit here had been extended to 4 keV using Mughabghab’s listed parameters in the Atlas [55]. By extending the fit to 4 keV, the RRR model was being applied to a region where the measurement was not completely resolved. Despite this, the RRR model recreated the measured transmission and capture yield reasonably well.

5.19 The thick-sample transmission measurement and evaluated transmissions are compared to the RPI extended RRR evaluation in black. The extension of the RRR caused some discrepancies between 3 and 4 keV, and no longer seemed to follow the grouped structure of the experimental data. The additional resonances from 2.5 to 4 keV also contribute enough cross section interference to reduce the potential scattering (and increase the transmission) at 2 keV.

5.20 The average total cross section for the 3 and 6 mm Ta measurements, along with the fitted theoretical total cross section in red. The fitted cross section comes from average resonance parameters fit in the energy regions 2→10 keV, 10→45 keV, and 45→120 keV.

5.21 The average capture cross section for the 1 and 2 mm Ta capture measurements, along with the fitted theoretical capture cross section in red. The fitted cross section comes from average resonance parameters fit in the energy regions 2→10 keV, 10→45 keV, and 45→120 keV.

5.22 The validation transmission measurement and evaluated transmissions compared the RPI evaluation with a multiple energy region fit of the URR. Fitting multiple energy regions in the URR was not seen to have a pronounced improvement over fitting the full energy range of experimental data.
5.23 The computed \( k_{\text{eff}} \) values \( C \) corresponding to the simulation of the TEX [25] benchmarks, with Ta cross sections reported in the ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 libraries, normalized to the computed \( k_{\text{eff}} \) value using the ENDF/B-VIII.0 evaluation of Ta: \( C_{\text{ENDF}} \). This comparison shows the impact of using the reported cross sections for Ta from different evaluated libraries. The difference in \( k_{\text{eff}} \) between JENDL-4.0 and ENDF/B-VIII.0 is \( \frac{\Delta k}{k} \approx 800 \text{ pcm or } 0.8\% \).

5.24 The computed \( k_{\text{eff}} \) values corresponding to the simulation of the TEX [25] benchmarks, with Ta cross sections reported in the ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 libraries (squares) \( C \), normalized to the computed \( k_{\text{eff}} \) value using the ENDF/B-VIII.0 evaluation of Ta, \( C_{\text{ENDF}} \). Also shown are the \( C/C_{\text{ENDF}} \) values for the URR and full RRR/URR RPI evaluation (circles). The difference in \( k_{\text{eff}} \) between JEFF-3.3 and the RPI “URR only” evaluation is approximately \( \frac{\Delta k}{k} \approx 200 \text{ pcm.} \) This difference highlights the importance of the URR cross section and resonance self shielding effect for criticality.

5.25 The sensitivity \( (\frac{\Delta k_{\text{eff}}}{k_{\text{eff}}} / \frac{\Delta \sigma_{\text{r}}}{\sigma_{\text{r}}}) \) of the computed \( k_{\text{eff}} \) value to the RPI evaluated capture (\( \sigma_{\text{c}} \)) and elastic scattering (\( \sigma_{\text{r}} \)) cross sections. The TEX benchmark is more sensitive to the capture cross section than the scattering cross section. It should be noted that \( \sigma_{\text{r}} \approx \sigma_{\text{c}} + \sigma_{\text{n}} \) for energies < 100 keV.

6.1 A compilation of thermal total cross section values compiled by Barry [88]. The horizontal solid and dashed red lines indicate the evaluated value published by Mughabghab in the Atlas of Neutron Resonances [55].

6.2 A compilation of thermal capture cross section values compiled by Barry [88]. The horizontal solid and dashed red lines indicate the evaluated value published by Mughabghab in the Atlas of Neutron Resonances [55].

6.3 Pb shield designed to reduce the room background measured on by the capture system by a factor of \( \approx 1.5-2x \).

D.1 The URR cross section for \(^{181}\text{Ta}\) as calculated by RECONR (red), PURR (black X’s), and FITACS (green triangles). The RECONR module gave a more reasonable cross section result when the set of inelastic widths for \( L=1 \), and \( J=2 \) were set to zero (blue). The inelastic cross section begins to compete at 7.2 keV, as indicated by the vertical dashed black line.

F.1 The values \( \frac{C_{\text{mon}}/C_{\text{det}}}{(C_{\text{mon}}/C_{\text{det}})} \) have been plotted as a function of cycle number for \(^{181}\text{Ta}\) and Open from the thick-sample transmission measurement. The dashed horizontal lines indicate the 2-sigma uncertainty for Ta (blue) and Open (red).
ACKNOWLEDGMENT

In 1895 Wilhelm Roentgen inadvertently discovered the x-ray while tinkering with a cathode ray tube near a fluorescent screen in his laboratory. Some unknown radiation had passed through cardboard surrounding the cathode ray tube and caused the screen to fluoresce. When later asked what he thought upon seeing the fluorescent effects of the x-rays on the screen, he replied, “I did not think; I investigated.” The culmination of my time as a Ph.D. candidate leads to this concise statement. Whether it was done with an electron accelerator, or with pen and paper, my advisors and colleagues in the RPI Nuclear Data group taught me to investigate and immediately dive into the work and seek the unknown which had caused the effect I’d observed.

The first person I must thank for the strengthening of my scientific understanding is my thesis advisor Dr. Yaron Danon, whose professional example has been indispensable. His propensity to seek the fundamental mechanisms of everything from computer programs to resonance theory is something I’d like to emulate in my professional life. I’d also like to thank Dr. Robert Block for his seemingly bottomless expertise on nuclear physics and neutron experiments, as well as his willingness to continue dispensing his wisdom to generations of nuclear engineers.

I’d like to thank my doctoral committee as well. Dr. Emily Liu’s class on nuclear physics broadened my understanding of nuclear structure, and Dr. Wei Ji’s class on neutron transport methods was an excellent exercise in combining programming and nuclear science. Dr. Hyun Gook Kang’s class on probabilistic risk assessment for nuclear reactors exposed me to an exciting aspect of nuclear engineering that I deeply respect, and hope can have an impact on nuclear reactor safety and reliability. I would like to thank Dr. Vladimir Sobes for the opportunity to work with him during an internship at Oak Ridge National Laboratory. His friendship and professional guidance were just as important to me as his instruction in methodical problem solving and resonance evaluation. I must also extend special thanks to Dr. Devin Barry, for his willingness to endure some of my most obtuse questions, as well as his impeccable guidance towards enlightening books, articles, and computer programs that enabled me to complete this dissertation.
I owe many thanks to the Linac Technicians Matt Gray, Larry Krusieski, Azeddine Kerdoun, Michael Bretti, and their fearless leader Peter Brand, without whom the neutrons would never fly. This work also would not have been possible without the support from Naval Nuclear Laboratory researchers: Brian Epping, Tim Trumbull, Greg Leinweber, Mike Rapp and John Burke.

I have to thank my colleagues at RPI for their support. Amanda Youmans, Kumar Mohindroo, and Hyun Jin Choun for the many chats over coffee and Thai food, my office mates Kemal Ramic and Adam Ney, and many other nuclear engineering students. Dr. Brian McDermott whose work I was able to build on. Dr. Adam Daskalakis for our conversations of graduate work. Dr. Ezekiel Blain for his expertise on nuclear physics and radiation detection, as well as his diligent review of my publications. Dr. Nicholas Thompson for his guidance on all aspects of graduate work and life. And to the many others who have shared in my experience.

I owe a huge debt of gratitude to my family, especially my father Tony Brown and mother Vonda Brown, who worked hard to ensure the success of their children. My mother taught me to enjoy reading which has been the cornerstone of my education. My father gave me a box of books on biology and zoology that initiated my love for science. This work is dedicated to them.

Finally I must thank my wife Lindsay whose hard work, brilliance, and kindness is an inspiration. Without her emotional support, this work would not have been possible.
ABSTRACT

$^{181}$Ta is a metal which is resistant to heat and chemical reactions, making it a desirable material for nuclear applications. For any material used in a nuclear application the neutron cross section must be well known, or large uncertainties will exist in the behavior of the application. There are discrepancies between the reported neutron cross section for $^{181}$Ta in evaluated libraries. Total cross section and capture cross section measurements must be consulted to resolve the discrepancies found in the evaluations. The data available to the public, however, is inadequate in some energy regions. The lack of high-resolution energy differential data, in the keV region specifically, motivated performing high-resolution measurements. In addition to reporting discrepant nuclear parameters for $^{181}$Ta, evaluated libraries also apply resolved and unresolved resonance treatments to different energy regions, which can result in different predicted cross sections and observables such as transmission.

Neutron transmission and neutron capture yield measurements were made at Rensselaer Polytechnic Institute to obtain energy differential cross section data that was not previously available to the nuclear engineering and physics communities. This was done in an effort to support a new evaluation. An important feature of this evaluation dataset is that the transmission and capture yield measurements were made with the same experimental conditions. As the $^{181}$Ta nucleus only allows elastic scattering and capture reactions (with any significant probability) in the energy range of interest (1 eV to 100 keV), this combined dataset constrains the cross sections of interest much better than separate measurements. This combined dataset is a valuable resource that can be used to improve the community’s understanding of how $^{181}$Ta reacts to neutrons from approximately 1 eV to 100 keV.

Following the evaluation of $^{181}$Ta it is important to validate the final evaluated cross section. This has commonly been done with integral benchmarks. It has been shown in this work that, in addition to the well-known integral benchmarks, an energy-differential thick-sample transmission measurement can be used to better validate evaluated data. This is shown by modeling transmission with a continuous energy Monte Carlo code. The transmission validation method developed in this work is designed specifically to probe the resonance self-shielding effect in the unresolved resonance region. This is a novel application of a thick-

xix
sample transmission measurement in the unresolved resonance region, which focuses on the self-shielding effect.

This energy-differential validation method is also capable of answering the question evaluators pose about the extension of the resolved resonance region. Often evaluators wish to extend the resolved resonance region beyond the energies where conventional wisdom would advise against it (because it is partially unresolved), in an effort to improve the cross section model for applications. The transmission validation method can be used to verify that the partially resolved cross section either does or does not properly predict transmission. It was found that the extension of the $^{181}$Ta resolved resonance region to 2.4 keV predicted the thick-sample transmission measurement well and did not negatively affect the integral benchmarks tested in this work. It was also found that the unresolved resonance parameters in the major evaluated libraries did not properly model the thick-sample transmission measurement. To address this, the separate combined set of transmission and capture yield measurements were evaluated in the resolved and unresolved resonance regions and compared to the thick-sample transmission measurement. Good agreement was found between the Rensselaer Polytechnic Institute evaluation and the transmission validation measurement. The thick-sample transmission measurement demonstrated here can be applied to other isotopes to validate URR evaluated parameters, ultimately enabling more accurate modeling of nuclear applications.
CHAPTER 1
INTRODUCTION

1.1 Nuclear Data

Nuclear interactions are described by semi-empirical models [1]; this means that the mathematical equations used to describe nuclear interactions require empirical values to complete the physical model. These vital empirical values are maintained in evaluated databases to be used by academia and industry. Typically, to be accepted into an evaluated database, the data must be verified by an evaluator to accurately represent physics and match corresponding experimental evidence given by benchmark experiments. The physics and nuclear engineering communities have created advanced tools to model and simulate systems, such as nuclear reactors [2], that depend on these nuclear data. With the advances of computer technology over the last few decades, modeling and simulation can carry out very high precision mathematical analysis and store enormous geometric detail of modeled systems. The result being that the uncertainty in the data used to complete the physical interaction model becomes a large fraction of the overall uncertainty in the modeled system. For this reason, it is important to maintain, update, and improve nuclear data evaluations.

In the United States, the National Nuclear Data Center (NNDC) maintains the ENDF/B [3] library which contains the necessary information to describe many nuclear interactions. One of the most important of these interactions is a neutron’s probability to interact with a nucleus, or neutron cross section. Neutron cross sections are important to many fields of engineering and physics including: nuclear power reactors, astrophysics [4], nuclear medicine [5], and many more.

The cross section of Ta in particular is important to applications as well. Tantalum is a metal which is chemically resistant [6] and has a high melting temperature of 3269°K [7]. The natural isotopic abundance of Ta is 99.99% $^{181}$Ta [8]. The natural properties of Ta make it a desirable material for the fabrication of anything that must survive an inhospitable environment, such as a reactor. Some of the uses of Ta that can be found in literature are: as test tubes and crucibles in the experimentation with molten actinides [9][10][11][12], the High Flux Isotope Reactor (HFIR) [13] at Oak Ridge National Laboratory (ORNL), and the Prometheus space reactor project [14].
If we follow the logic found in Duderstadt [15], neutron cross section can be defined as follows: we consider a beam of neutrons all traveling at the same speed and direction. The direction of the neutron beam is orthogonal to the surface of the material with which it collides. This is depicted in Fig. 1.1.

**Figure 1.1:** A diagram to depict how cross section is defined by beam intensity $I$, atomic density $N$, and sample thickness $dx$. The arrows represent the direction orthogonal to the face of the target sample.

Assuming that this material is one atomic layer thick (so none of the nuclei are shielded by any of the other nuclei), we would expect the reaction rate $R$ of the neutrons with the target nuclei to be proportional to the intensity of the neutron beam $I$, and the atomic or nuclear areal density $n = N \cdot dx$. In this notation $N$ is the atomic density, and $dx$ is the sample thickness. The constant of proportionality is called the cross section $\sigma$, as described in Eq. 1.1.

$$R \left[ \frac{\#}{cm^2 s} \right] = \sigma \left[ cm^2 \right] I \left[ \frac{\#}{cm^2 s} \right] n \left[ \frac{\#}{cm^2} \right] \quad (1.1)$$

The microscopic neutron cross section can then be thought of as the area that each nucleus in the target presents to the neutron beam. If neutrons and nuclei behaved as classical particles and the nuclear radius was about $10^{-12}$ cm, it would be natural to assume that this cross sectional area would be on the order of about $10^{-24}$ cm$^2$ or 1 barn (sometimes simply 1 b). However, due to the wave-like properties of the neutron and nucleus, the cross section can cross many orders of magnitude. An example of an evaluated total cross section for $^{181}$Ta [16] is shown in Fig. 1.2.
Figure 1.2: The total cross section for $^{181}$Ta as evaluated in JEFF-3.3 [16]. RRR represents the resolved resonance region, URR the unresolved resonance region, and Continuum represents the continuum region where the cross section changes slowly as a function of energy.

The total cross section $\sigma_t$ is the sum of the reaction cross sections $\sigma_t = \sigma_\gamma + \sigma_s + \sigma_f + (\ldots)$ possible for a given isotope. If the center-of-mass (COM) energy of an incident neutron and target nucleus pair matches an energy level $\lambda$ in that nucleus, the probability for interaction is greatly increased producing what are called resonances. This is the cause of the sharp peaks in cross section seen in Fig. 1.2. A resonance can be described by its location in energy space $E_\lambda$, and its total width $\Gamma_t = \Gamma_n + \Gamma_\gamma + \Gamma_f + (\ldots)$, which is a sum of the neutron width, radiative capture width, fission width, etc. For neutron cross sections the neutron width $\Gamma_n$ has an energy dependence $\propto \sqrt{E}$ (s-waves)[15]. Cross section is often classified into several different regions of energy, each with a corresponding semi-empirical model that best describes the physics of the region; a description of these regions can also be found in Ref. [17]. There is a resolved resonance region (RRR) model for the lower energy region, the unresolved resonance region (URR) model for energies just beyond the RRR, and the fast or continuum region optical model for where the cross section varies slowly as a function of energy; these are labeled in Fig. 1.2.

In the RRR $\Gamma_t < D$, where D is the average distance in energy between levels of the nucleus; this allows resonances to be clearly defined in energy space. In energy differential
measurements of cross section, however, there is always uncertainty in neutron energy $\Delta E$. Any measurement of cross section is measuring an effective average value of the true cross section in $\Delta E$. If the uncertainty in the energy of the neutron $\Delta E > D$, the resonances can no longer be resolved. We define the URR as the energy region where the true cross section has separated resonances, but no experimental data exists to resolve them. As neutron energy increases, eventually the increase in $\Gamma_n$ causes the inequality $\Gamma_t > D$ to become true; the resonances significantly overlap with each other causing the true cross section to vary slowly as a function of energy in the continuum region.

1.2 Time-of-Flight Measurements

Neutron time-of-flight (TOF) measurements are the principle way to experimentally determine energy-differential neutron cross section. TOF measurements require high precision knowledge of the time at which a free neutron is born $t_0$ and the time bin $t_i$ at which it was detected. The difference of these times $TOF = t_i - t_0$, along with the knowledge of a fixed flight path length $L$, can be used to calculate the kinetic energy of the neutron $E$. The special relativistic equation can be used as in Eq. 1.2, where $m_n$ is the neutron mass, $c$ is the speed of light, and $E$ is the energy of the neutron.

$$E = m_n c^2 \left( \frac{1}{\sqrt{1 - \left(\frac{L}{t_i - t_0}\right)^2}} - 1 \right) \quad (1.2)$$

For neutron energies where relativistic effects are very small (for example $E \approx < 1$ MeV) the energy of the neutron can be calculated by the familiar kinetic energy relation Eq. 1.3 where $k \approx 72.29 \left(\frac{\text{eV} \cdot \mu\text{s}}{m}\right)$. The error in the non-relativistic energy equation is plotted as a function of energy in Fig. 1.3.

$$E = \frac{1}{2}mv^2 = \frac{1}{2} m \left( \frac{L}{t_i - t_0} \right)^2 = \left( \frac{k \cdot L}{t_i - t_0} \right)^2 \quad (1.3)$$
Figure 1.3: The percent error in the non-relativistic approximation for kinetic energy as a function of energy.

TOF experiments described in this PhD work include neutron transmission and radiative neutron capture yield. TOF measurements at RPI employ a pulsed neutron source, where the time of neutron evaporation from the neutron producing target can be accurately recorded. Pulses occur at a constant rate in time such that each burst of neutrons is separated in TOF enough to cover the desired energy range.

1.2.1 Neutron Transmission

Neutron transmission is a conceptually simple experiment where the intensity of a neutron beam \( I(x) \) is measured with and without a sample in the beam. The idea as described in Eq. 1.4 is that \( I(x) \) changes proportionally to \( N\sigma_t \), where \( \sigma_t \) (cm\(^2\)) is the total cross section, \( N \) (1/cm\(^3\)) is the atomic density, and \( x \) (cm) is some distance traveled in the material. From these definitions, we can define a differential equation.

\[
dI(x) = N\sigma_t I(x) dx \quad \rightarrow \quad \frac{dI}{dx} = N\sigma_t I
\]

The solution of that differential equation is given by Eq. 1.5 which describes the beam intensity as a function of \( x \), for some initial beam intensity \( I_0 \).
\[ I(x) = I_0 e^{-N\sigma_t x} \] (1.5)

The measurement of beam intensity without a sample in the beam is taken as \( I_0 \), the intensity measured with some sample of thickness \( x \) in the beam is \( I(x) \). With these two measurements, the neutron transmission can be calculated by their ratio at each TOF (or neutron energy \( E \)), as shown in Eq. 1.6. In a real experiment measured neutron intensities include background associated with the measured \( I(x) \) and \( I_0 \); this has been discussed in detail in §4.1. Solving the equation for the total cross section \( \sigma_t \) results in Eq. 1.7. A diagram of a typical neutron transmission experiment is shown in Fig. 1.4.

\[ T(E) = \frac{I(x)}{I_0} = e^{-N\sigma_t(E)x} \] (1.6)

\[ \sigma_t(E) = -\frac{1}{N_x} \ln[T(E)] \] (1.7)

---

Figure 1.4: A diagram of a typical TOF transmission experiment. Electron pulses are accelerated toward a neutron-producing target. The pulsed neutron emission is collimated towards a sample, collimated after the sample, and recorded by a neutron detector at a fixed flight path length.

The diagram illustrates a pulse of electrons directed toward a neutron producing target. When electrons collide with the electric field of atoms in the target Bremsstrahlung radiation is emitted and interacts with the nuclei of the target in a \( (\gamma, n) \) reaction. This causes an evaporation of free neutrons from the target as a “white” source, meaning there are neutrons
of many different energies. Often there are moderators such as water in the target; neutrons collide with water molecules and lose energy softening the overall neutron spectrum. The target emits neutrons in all directions, but only those which are emitted in the direction of the sample and detector are recorded. This neutron beam is collimated before and after the sample to achieve good geometry with the detector. Neutrons are emitted in short pulses, travel the $L$ of fixed length, and finally are recorded by a neutron detector at the end of the flight path in a time bin $t_i$. The time of neutron emission, $t_0$, is equated to the time of Bremsstrahlung radiation emission, often called the “gamma flash”.

### 1.2.2 Neutron Radiative Capture

Radiative neutron capture experiments involve measuring a neutron beam rate that is incident on a sample $R_n$, and the rate of neutron captures in that sample $C_\gamma$. The ratio of these two quantities is called the radiative capture yield $Y$, which is shown in Eq. 1.8.

$$Y = \frac{C_\gamma}{R_n} \quad (1.8)$$

In Eq. 1.8 we have ignored the background that is associated with the measurements of $C_\gamma$ and $R_n$, these will be discussed further in §4.2. The theoretical yield is related to the capture cross section in Eq. 1.9, where we integrate the probability for transmission over the sample thickness $x$ multiplied by the probability per unit path length for capture $N\sigma_\gamma$.

$$Y(E) = N\sigma_\gamma(E) \int_0^x e^{-N\sigma_t(E)x'} dx' \quad (1.9)$$

Finally, we come to Eq. 1.10, which is the relation of $Y$ to the product of two probabilities: the probability to interact over thickness $x$ given by $(1 - e^{-N\sigma_t x})$ and the probability that the interaction be a capture reaction is given in the leading fraction $\frac{\sigma_\gamma}{\sigma_t}$. The energy dependence notation in Eq. 1.10 is dropped for simplicity. It should be noted here that Eq. 1.10 is for the primary yield, that is, it ignores the possibility of multiple scattering where the neutron first scatters and then captures in the sample. Solving for the capture cross section $\sigma_\gamma$ we come to Eq. 1.11. If we assume that the sample is thin ($Nx << 1$) we can approximate the term in parentheses with a Taylor series expansion. A typical yield experimental setup is shown in Fig. 1.5.
\[
Y = \frac{\sigma_{\gamma}}{\sigma_t} \left(1 - e^{-N\sigma_t x}\right)
\]  
(1.10)

\[
\sigma_{\gamma} = \frac{Y \sigma_t}{(1 - e^{-N\sigma_t x})} \approx \frac{Y \varphi_i}{N_x \varphi_i} = \frac{Y}{N_x}
\]  
(1.11)

Figure 1.5: A diagram of a typical TOF radiative capture yield experiment. The pulsed neutron beam is collimated towards a sample, which may capture a neutron and emit a cascade of capture photons. Capture photons emitted in the direction of the photon detector are recorded as a function of TOF.

Same as with the transmission experiment, a pulsed neutron source is collimated towards the sample of interest. In capture measurements, however, the rate of neutrons reaching the sample is recorded and the rate of neutrons capturing in the sample is recorded. The rate at which neutrons are striking the sample was determined by a reference sample, which will be discussed further in §4.2. The capture measurement setup is illustrated in Fig. 1.5 where an incident neutron is captured by a nucleus and the nucleus de-excites by emitting a cascade of photons. The emitted photon which can be recorded is represented by the oscillating line traveling from the sample to a detector.

1.3 Nuclear Data Evaluation

Over all energies, neutron cross section is represented by semi-empirical models which require experimentally determined parameters. In order to model the RRR the \(\mathfrak{R}\)-matrix theory was formalized by Lane and Thomas [18]. This model requires resonance parameters
such as $E_\lambda$, $\Gamma_n$, $\Gamma_\gamma$, etc. Modeling cross section in the URR most often employs the average $\mathfrak{R}$-matrix and Hauser-Feshbach theory [19], which uses parameters such as the strength function $S_l$, average radiation width $\langle \Gamma_\gamma \rangle$, etc. which have been discussed further in §2.

These parameters are extracted from experimental data when possible or estimated by theory if none are available. At times, multiple sets of experimental data are available to describe the same interaction. The process of evaluation is choosing how to weight the data that are available, and how to model the cross sections for all of the energy regimes. The resulting evaluated cross sections can then be verified by using them to calculate how simulated benchmark systems like reactors compare to equivalent experimental systems.

1.4 Tantalum: Background and Motivation

Evaluated nuclear data libraries ENDF/B-VIII.0 [3], JEFF-3.3 [20], and JENDL-4.0 [21] each define different energy boundaries for the RRR, URR, and high energy continuum region. JEFF-3.3, CENDL-3.1 [22], and ROSFOND-2010 [23] all state that they obtained RRR and URR parameters from JENDL-3.3 [24]; to avoid redundancy only JEFF-3.3, JENDL-4.0, and ENDF/B-VIII.0 are discussed. JEFF-3.3 and JENDL-4.0 are almost identical evaluations for $^{181}$Ta but have small differences in the URR. The different choices evaluators made in models and storage methods for neutron cross section data served to highlight the impact those choices had on final applications. ENDF/B-VIII.0 and JEFF-3.3/JENDL-4.0 end the RRR at 330 eV and 2400 eV respectively. ENDF/B-VIII.0 and JEFF-3.3/JENDL-4.0 end the URR at 5 keV and 100 keV respectively. This is shown in Table 1.1 and the cross section is plotted in Fig. 1.6.

Table 1.1: The boundaries for the end of the RRR and URR for each of the libraries discussed during this work.

<table>
<thead>
<tr>
<th>Evaluation</th>
<th>End of RRR</th>
<th>End of URR</th>
</tr>
</thead>
<tbody>
<tr>
<td>ENDF/B-VIII.0</td>
<td>330 eV</td>
<td>5 keV</td>
</tr>
<tr>
<td>JEFF-3.3</td>
<td>2.4 keV</td>
<td>100 keV</td>
</tr>
<tr>
<td>JENDL-4.0</td>
<td>2.4 keV</td>
<td>100 keV</td>
</tr>
</tbody>
</table>

The evaluated $^{181}$Ta total cross section for the mentioned libraries are shown in Fig. 1.6. The differences in cross section evaluations result in discrepancies between libraries in, for example, calculated neutron transmission and capture rate, as well as the proposed Thermal/Epithermal eXperiments (TEX) benchmarks [25]. The TEX benchmarks have not yet
Figure 1.6: Shown above are the evaluated total cross sections from the JEFF-3.3, ENDF/B-VIII.0, and JENDL-4.0 libraries. The energy regions (RRR: 1-2400 eV, URR: 2.4-100 keV, Continuum: 0.1-10 MeV) are labeled by the model which is used to represent them in JEFF-3.3, and JENDL-4.0. It can be seen that ENDF/B-VIII.0 deviates significantly from the other evaluations: ENDF/B-VIII.0 sets the end of the RRR at 330 eV, and ends the URR model at 5 keV. The JEFF-3.3 and JENDL-4.0 libraries are nearly identical.

been published, but preliminary reports have been released on these criticality benchmarks that focus on epithermal and intermediate neutron energies. A preliminary model of one of the proposed benchmark experiments (Exp. 10) was used to demonstrate the effects that different evaluations have on an integral system.

Using the Monte Carlo code MCNP 6.1 [26] to simulate the TEX criticality experiment provides insight on how the cross sections will affect the neutron multiplication factor $k_{eff}$. For each criticality simulation in MCNP 6.1, all isotopes used ENDF/B-VIII.0 libraries and the $^{181}$Ta cross section is changed to the library of interest. As there is no experimental value for $k_{eff}$ (as the TEX benchmarks have not been completed), each simulated $k_{eff}$ was compared to ENDF/B-VIII.0 and the computed values $C$ over the computed value for ENDF/B-VIII.0 $C_{ENDF}$ are plotted for each $^{181}$Ta evaluation in Fig. 1.7. In this way,
Figure 1.7: The normalized computed neutron multiplication factor $C/C_{ENDF}$ for ENDF/B-VIII.0, JEFF-3.3 and JENDL-4.0 cross section evaluations of Ta employed in the preliminary TEX benchmarks [25]. For each criticality calculation, only the cross section library for $^{181}$Ta is changed. This shows the impact of the different evaluated $^{181}$Ta cross sections on epithermal criticality applications.

values provide a measure on how sensitive the benchmark is to the $^{181}$Ta cross section, though they don’t provide information on which evaluation is closer to an experimental $k_{eff}$.

It can be seen in Fig. 1.7 that the calculated criticality can change by as much as $\approx 0.8\%$ by changing the evaluated library for $^{181}$Ta. To determine the validity of any of the evaluated cross sections, they must be compared to the available cross section measurements for $^{181}$Ta. Upon finding the available experimental data to be unavailable or of inadequate resolution for the intermediate energy region, it was determined new measurements should be made. In an effort to improve the neutron cross section evaluation of $^{181}$Ta, high resolution TOF measurements of neutron transmission and capture yield were made at the Gaerttner Laboratory Electron Linear Accelerator (Linac) at Rensselaer Polytechnic Institute (RPI).

The history of nuclear TOF data for $^{181}$Ta goes back as early as 1947 in which the Havens et. al measurement produced resonance parameters between the energies of 4.1 and 37 eV [27]. Most of the work relevant to current evaluations, however, is listed in the information file portion of the standard ENDF format [28], which can be found at the very beginning of the ENDF file. ENDF/B-VIII.0 references the work of S.F. Mughabghab and Gader [29] from 1973 for the RRR parameters and Macklin et. al [30] from 1984 for the
capture cross section in the URR. An assumption of $\Gamma_\gamma = 0.059$ eV was made for resonances of unknown radiation width. JEFF-3.3 uses the RRR and URR data from JENDL-3.3. JENDL-4.0 takes the RRR parameters from JENDL-3.3 and determines URR parameters to reproduce measured capture cross sections from Macklin et. al and Yamamuro et. al. [31]. JENDL-3.3 references the same works of Mughabghab and Macklin et. al, with the addition of private communication with Tsubone et. al. Tsubone et. al published an article on neutron transmission of $^{181}$Ta between 0.1 and 4.3 keV [32] in the same year of 1987 that the JENDL-3.3 evaluation was done. Both the Macklin ($L = 40$ m, pulse width = 2.7 ns) and Tsubone ($L = 200$ m, pulse width = 30 ns) measurements were capable of recording high resolution data, but the published Macklin data were grouped heavily and the Tsubone transmission dataset is not publicly available, preventing further analysis. The energy regions and measurement types are shown in graphical form in Fig. 1.8.

Figure 1.8: Energy regions covered by referenced works listed by evaluators in the information file portion of each ENDF file for $^{181}$Ta, and the corresponding data type. The dashed vertical lines delineate the beginning and end of the shared JEFF-3.3/JENDL-4.0 RRR.

Also shown in Fig. 1.8 are the energy regions covered by new high resolution measurement made by McDermott et. al [33] and the measurements made for the present Ph.D. work (Brown). Additional previous measurements are available online from the International Atomic Energy Agency (IAEA) EXFOR [34], a database for experimental nuclear reaction data compiled by the Nuclear Reaction Data Centres (NRDC). These data include neutron total and capture cross sections and, for some isotopes, covariance information. Fig. 1.9
Figure 1.9: A representative selection of available total and capture cross section experimental data for $^{181}$Ta from the IAEA EXFOR [34] database. Data sources are from Refs. [31],[33],[35],[30],[36],[37],[38],[39],[40],[41].

shows an example of the data available to researchers for neutron total and capture cross sections for $^{181}$Ta.

The energy region of interest for this work was the region from 300 eV to 100 keV. This region is where most of the discrepancies in the cross sections of evaluated libraries are found for $^{181}$Ta. Among the information found in EXFOR, none of the measurements were capable of resolving resonances above 1 keV, except a transmission measurement in EXFOR by Harvey [34]. The Harvey data are from a measurement with excellent energy resolution (80 m flight path, 20 ns burst width) for one sample thickness, the other was measured at a shorter flight path and too thin to observe many of the resonances above 100 eV. The Harvey measurements are included in the analysis of this Ph.D. work, and plotted in Fig. 1.10. No capture measurements were found which resolved resonances in the energy region of interest.

In a TOF experiment by Thompson [42] at the RPI Linac, a lead slowing-down spectrometer (LSDS) was employed to measure capture cross sections of several materials including a thin (0.010") sample of $^{181}$Ta. The LSDS is designed to receive a pulse of neutrons
in its center and scatter them many times, which allows a sample placed within it many opportunities to capture. This results in a very high count rate and relatively low energy resolution, allowing for statistically accurate measurements that cover large energy ranges. As demonstrated by Thompson, using the ENDF/B-VII.1, JEFF-3.2, and JENDL-4.0 libraries to simulate capture rates in the LSDS resulted in discrepancies amongst the libraries. Additionally, in the region where JEFF-3.2 and JENDL-4.0 have extended the RRR to 2.5 keV, the simulations match the $^{181}$Ta capture measurement better than ENDF/B-VII.1 (URR from 0.33-5 keV). This is shown in Fig. 1.11. It should be noted that the resonance parameters listed in ENDF/B-VIII.0 have largely remained unchanged since the 1973 publication by Mughabghab [29], and the JEFF-3.3 RRR is reliant entirely on the JENDL-3.3 evaluation. These discrepancies and the lack of high-resolution data available to the community warrant new measurements and evaluation.
1.5 Validation Transmission

In the field of nuclear engineering, benchmarks are typically defined as trusted experiments which depend on neutron cross section. In order to accurately model the behavior of the benchmark experiment, the cross sections used to model the experiment must also be accurate. Evaluators use benchmark experiments to validate the cross sections that they provide in evaluated libraries. Traditionally integral benchmarks are used to verify the accuracy of evaluated neutron cross section data. Integral benchmarks, as the name suggests, are benchmark experiments which depend on the sum of cross sections across many different energies and angles. As a result, even if cross sections are inaccurate at some energies, if the sum of the weighted cross section (the weighting depends on the integral system) is correct the benchmark will be met.

In addition to these commonly used benchmarks, high energy shielding benchmarks have been used to verify the high energy cross section, as demonstrated by Yamano [43]. Often these shielding benchmarks compare the flux measured from a source within a thick spherical shell, such as that by Jansky [44] or Simakov [45]. To match the spherical shell
benchmarks, many different aspects of the cross section must be accurate, such as the angular distributions and the ratio of scattering and capture probabilities $\sigma_\gamma/\sigma_s$. Other previously adopted benchmarks are specifically designed to measure the angular distributions of the high energy cross section such as those by Malaviya [46] and Murata [47].

To measure the total cross section without additional confounding effects, however, a simpler benchmark termed the broomstick transmission has been employed. This type of measurement uses a sample that is shaped as a slender cylinder, similar to a broomstick. The cylinder axis is aligned in the direction of a neutron beam which allows any scattered neutrons to exit the detection system, and the transmitted neutron count rate is recorded. Examples of broomstick transmissions are those by Maerker [48][49] at Oak Ridge National Laboratory (ORNL). An imitation of the experimental design from the Maerker report is shown in Fig. 1.12.

![Experimental design of the 1972 Fe broomstick transmission by Maerker [48]](image)

Figure 1.12: Experimental design of the 1972 Fe broomstick transmission by Maerker [48]. Neutrons are emitted from a reactor, collimated towards a cylindrical sample of Fe, and finally collimated further towards a proton recoil detector. The energy of the transmitted neutrons is calculated by the energy deposited from a proton recoil, which has lower energy accuracy than the TOF method. (Note: Not to scale.)

Part of the Murata benchmarks [47] for Cu and W include a high energy transmission measurement. The Murata measurement uses a DT source to generate neutrons and measures the TOF of the neutrons across an 8 m flight path. The experimental setup for the
Murata measurement is shown in Fig. 1.13, which is an approximation of the geometry seen in Murata’s publication.

![Diagram of Murata's measurement setup](image)

**Figure 1.13:** The approximate geometry from the 2001 publication by Murata on high energy transmission and angular benchmarks using TOF techniques [47]. The geometry is highly collimated with a short flight path of 8 m. (Note: Not to scale.)

In the present Ph.D. work, a new tool for evaluators is proposed for validating the URR: a differential thick sample transmission measurement. Among the previously mentioned benchmarks, this new validation transmission method most closely resembles the transmission measurements of Murata but focuses on the URR cross section. The RPI Linac and neutron production target are used as a source of neutrons, and a well collimated transmission of a thick sample is measured with a configuration like that seen in Fig. 1.14. The RPI measurement places the neutron producing target off axis and reflects neutrons into the beam path with a polyethylene disc. This is done to reduce the in-beam photon background originating from the electron induced Bremsstrahlung interaction and Ta decays.
The purpose of the thick \((n\sigma_t > 1)\) sample transmission measurement in this work is to validate the methods and parameters used to model cross section in the URR, expanding the capabilities of evaluators. The thickness of the sample chosen in this work (12 mm) was designed to optimize the uncertainty in transmission and resonance self-shielding. Methods which will be discussed include the choice evaluators make of where the boundary between resolved and unresolved resonances should lie, whether multiple sets of average parameters should describe different regions of the URR, and whether the URR should be represented by smooth cross section with underlying structure and subsequently have a fluctuation correction applied. The value of this novel validation transmission method is that it can be used to supplement integral benchmarks with more detailed information on the energy differential cross section, specifically in the URR where resonance self-shielding is important.
CHAPTER 2
NEUTRON-NUCLEUS INTERACTIONS

In order to understand the implications of the RRR and URR models used in data evaluation and applications, a brief discussion of the theory for the RRR and URR are presented here. As the resonance evaluation tool SAMMY [51] is used for much of the work presented here, the discussion will focus on how the various cross section models have been implemented in the SAMMY program.

2.1 RRR

In the RRR isolated resonances can be described with a set of parameters by the Breit Wigner formula [52]. An example of the Breit Wigner formula for capture cross section is given in Eq. 2.1.

$$\sigma_\gamma(E) = \sigma_0 \frac{\Gamma_\gamma}{\Gamma} \sqrt{\frac{E_\lambda}{E}} \frac{\Gamma^2}{\Gamma^2 + 4(E - E_\lambda)^2}, \quad \sigma_0 = 4\pi \lambda_0^2 \frac{\Gamma_n}{\Gamma}$$ (2.1)

Here $\lambda_0 = h/\sqrt{2m_nE_\lambda}$ is the reduced neutron wavelength, a quantity defined for each resonance $\lambda$. The variable $h$ is Planck’s constant divided by $2\pi$, and $m_n$ is the neutron mass. The total width $\Gamma = \Gamma_n + \Gamma_\gamma + ...$ is the sum of all partial widths, e.g., the neutron width and capture width are shown here. The resonance peak is located at approximately $E_\lambda$ and the incident neutron energy is $E$. The statistical factor $g = \frac{2J+1}{(2l+1)(2I+1)}$, where the intrinsic spin value of the neutron $i = 1/2$ and the intrinsic spin value of the target nucleus $I$ is an integer or half integer specific to that isotope with $+$ or $-$ parity. The vector addition of the channel spin $\vec{s}$, along with orbital angular momentum $\vec{l}$, gives the total angular momentum in units of $h$ for $\vec{J}$. The possible values for $\vec{J}$ and $\vec{s}$ are listed in Eqs. 2.2 and 2.3.

$$\vec{J} = \vec{l} + \vec{s}, \quad |l - s| \leq J \leq l + s$$ (2.2)

$$\vec{s} = \vec{l} + \vec{i}, \quad |I - i| \leq s \leq I + i$$ (2.3)
Figure 2.1: Diagram of the total, capture, and scattering cross section as computed by the Breit Wigner formula for the first resonance of $^{181}$Ta at energy $E_\lambda = 4.28$ eV.

The scattering cross section is shown in Eq. 2.4, where $R$ is introduced as the scattering radius. The first term in brackets corresponds to the resonance scattering, the second term in brackets is for the interference between resonance and potential scattering which produces the reduction in the cross section for energies just below $E_\lambda$. The last term in Eq. 2.4 is the potential scattering, which is simply the cross sectional area a sphere with radius $R$ would present to a neutron. The total cross section is the sum of the partial cross sections, e.g., here we have $\sigma_t = \sigma_s + \sigma_\gamma$. An example of the total cross section $\sigma_t$ as computed using the Breit Wigner formula is plotted in Fig. 2.1, along with the capture cross section $\sigma_\gamma$ and scattering cross section $\sigma_s$.

$$\sigma_s(E) = \sigma_0 \frac{1}{1 + \frac{4}{\Gamma^2} (E - E_\lambda)^2} \left[ \frac{\Gamma_n}{\Gamma} \sqrt{\frac{E_\lambda}{E}} + \frac{4R(E - E_\lambda)}{\chi_0 \Gamma} \right] + 4\pi R^2 \quad (2.4)$$

The Breit Wigner formula is often acceptable if resonances are well separated in energy space. Resonances of the same $J^\pi$ ($J$ is defined in 2.2 and $\pi$ as a superscript here indicates the parity of the neutron-nucleus interaction) may interfere with each other, however, and to properly model resonances with interference a more general mathematical model of resonance...
theory must be applied. $\mathcal{R}$-matrix theory, formalized by Lane and Thomas [18], is the preferred method to model resonances rigorously with interference effects. A brief description of $\mathcal{R}$-matrix theory, which is a phenomenological description of what is observed in cross sections, is given below. With notation as found in Ref. [17], the total cross section $\sigma_c$ for an entrance channel $c$ is the sum of partial cross sections $\sigma_{cc'}$ for exit channels $c'$ as shown in Eq. 2.5.

$$\sigma_c = \sum_{c'} \sigma_{cc'} = 2\pi \bar{\lambda}_c^2 g_c (1 - \text{Re}(U_{cc})) \tag{2.5}$$

The reaction channels $c, c' \equiv \{\alpha Jls, \alpha' J'l's'\}$ where $\alpha$ describes a reaction pair (e.g. $^{181}$Ta + n), $J$ is the total angular momentum, $l$ is the orbital angular momentum, and $s$ is the channel spin angular momentum. In Eq. 2.5 we use $\text{Re}(U_{cc})$ to indicate the real parts of the scattering matrix $U_{cc}$. The collision matrix $U_{cc}$ can be calculated using Eq. 2.6 and taking the $cc'$th element of the $U_{cc'}$ matrix.

$$U_{cc'} = e^{-i(\phi_c + \phi_c')} P_{cc'}^{1/2} \{ [1 - \mathbf{R}(\mathbf{L} - \mathbf{B})]^{-1} [1 - \mathbf{R}(\mathbf{L}^* - \mathbf{B})] \}_{cc'} P_{cc'}^{-1/2} \tag{2.6}$$

$$U_{cc'} = e^{-i(\phi_c + \phi_c')} \{ \delta_{cc'} + 2i P_{cc'}^{1/2} [(1 - \mathbf{RL}^0)^{-1} \mathbf{R}]_{cc'} P_{cc'}^{1/2} \} \tag{2.7}$$

In this notation variables in bold represent matrices and the same variables with subscripts provide additional information about the dimensions of the matrices. $L_{cc'}^0 \equiv L_{cc'} - B_{cc'} \equiv S_c - B_c + iP_c$, where $L_{cc'}$ are the logarithmic derivatives of the wave function, and $B_{cc'}$ are the logarithmic derivatives at the channel radius $a_c$. $P_c$ are the penetrabilities for the given channels, $S_c$ are the shift factors, and $\phi_c$ are the potential scattering phase shifts. The matrix $\mathbf{R}$ (the namesake of $\mathcal{R}$-matrix theory) is given by Eq. 2.8, where the $\gamma_{\lambda}$ values represent probability amplitudes for decay, $E_\lambda$ is the resonance energy, and $E$ is the energy of the incident particle (neutron). In the ENDF/B format most libraries use, the listed resonance widths are $\Gamma_{cc'} = 2\bar{\lambda}_c^2 \gamma_{cc'}^2$.

$$R_{cc'} = \sum_{\lambda} \frac{\gamma_{\lambda c} \gamma_{\lambda c'}}{E_\lambda - E} \tag{2.8}$$

It is common to use the Reich Moore approximation [53] to represent the $\mathcal{R}$-matrix where the interference of photon channels is ignored. For this approximation we replace the eigenvalue
\( E_\lambda \) with \( E_\lambda - i \gamma^2_{\lambda \gamma} \) and channels \( c, c' \) only represent the non-photon channels \((c, c' \notin \gamma)\), as shown in Eq. 2.9.

\[ R_{cc'} \approx \sum_\lambda \frac{\gamma_{\lambda c} \gamma_{\lambda c'}}{E_\lambda - E - i \gamma^2_{\lambda \gamma}} \quad (c, c' \notin \gamma) \quad (2.9) \]

SAMY 8.1 in default operation uses the Reich Moore approximation to the \( \mathcal{M} \)-matrix theory. The work presented here uses this formalism as well as an additional term \( R_c^{\text{ext}} \) added to the \( \mathbf{R} \) to account for contributions from distant levels. There are nuclear levels below the neutron binding energy \((E < 0)\) which cannot be observed and resonances above the measured RRR which cannot be resolved. These levels make a constant or slowly varying contribution to the cross section, and must be accounted for when fitting the RRR. The \( R_c^{\text{ext}} \) term is added to the \( \mathbf{R} \) as shown in Eq. 2.10.

\[ R_{cc'} = \sum_\lambda \frac{\gamma_{\lambda c} \gamma_{\lambda c'}}{E_\lambda - E - i \gamma^2_{\lambda \gamma}} + R_c^{\text{ext}} \delta_{cc'} \quad (2.10) \]

The \( R_c^{\text{ext}} \) term can be represented in SAMMY 8.1 as a constant or energy dependent function which can be fitted to the observed data. Fröhner and Bouland provide instruction in determining the \( R_c^{\text{ext}} \) term [54], which is defined in Eq. 2.11 as the difference between the sum of all levels and the observed levels \( \lambda \in \Lambda \), where \( \Lambda \) is the set of all observed levels.

\[ R_c^{\text{ext}} = \left( \sum_\lambda - \sum_{\lambda=1}^\Lambda \right) \frac{\gamma_{\lambda c} \gamma_{\lambda c'}}{E_\lambda - E - i \gamma^2_{\lambda \gamma}} \quad (2.11) \]

Fröhner then approximates the summation as an integral of continuous variables in the space of \( E' \) as seen in Eq. 2.12, where the energy interval of observed levels is \( I \) and the midpoint of the energy interval \( I \) is \( \bar{E} \).

\[ R_c^{\text{ext}} \approx \left( \int_{-\infty}^{\infty} \int_{E-1/2}^{E+1/2} \frac{dE'}{E_c} \langle \gamma_{c'} \gamma_c \rangle \frac{E' - E + i \langle \Gamma_\gamma \rangle / 2}{(E' - E)^2 + \langle \Gamma_\gamma \rangle^2 / 4} \right) \quad (2.12) \]

The assumption was made that, in the approximation of the integral where \( \langle \Gamma_\gamma \rangle \) is the average radiation width, the reciprocal value \( 1/D_c \) was the appropriate level density. If we further assume that \( (E' - E)^2 \gg \langle \Gamma_\gamma \rangle^2 / 4 \), that the off diagonal elements of \( \langle \gamma_{c'} \gamma_c \rangle \) are zero, and neglect the weak variation of the pole strength \( s_c \) over the integral, we can define the external contributions as shown in Eq. 2.13. The pole strength \( s_c \) is defined in Eq. 2.14, and
the distant level parameter \( R^\infty_c \) is defined in Eq. 2.15 where the integration is taken over all space \( E' \) excluding a infinitesimal space \( \epsilon \) around the singularity at \( E' = E \).

\[
R^{\text{ext}}_c = \left[ R^\infty_c (E) + 2s_c (\bar{E}) \right] \left\{ \text{arctanh} \left( \frac{E - \bar{E}}{I/2} \right) + i \frac{\langle \Gamma_\gamma \rangle}{I} \left[ 1 - \left( \frac{E - \bar{E}}{I/1} \right)^2 \right]^{-1} \right\} \delta_{cc'} \tag{2.13}
\]

\[
s_c (E) = \frac{\langle \gamma_c \rangle^2}{D_c} \tag{2.14}
\]

\[
R^\infty_c = \left( \int_{-\infty}^{E' - \epsilon/2} + \int_{E' + \epsilon/2}^{\infty} \right) \frac{s_c (E')}{E' - E} dE' \tag{2.15}
\]

The equations presented here to describe \( R^{\text{ext}}_c \) can be related back to the measured quantities of \( E_\lambda, \Gamma_\gamma, \) and \( \Gamma_{\lambda,n} \) with the following analysis. The pole strength can be defined by Eq. 2.16, where the strength function \( \tilde{S}_l \) is introduced as in Ref. [54].

\[
s_c = \frac{\tilde{S}_l}{2k_c a_c \sqrt{1[eV]/E}} \tag{2.16}
\]

In Eq. 2.16 the \([eV]\) is to indicate that the term inside the radical is unitless, it is not a variable. The wave number is represented by \( k_c = \sqrt{2m_a E A}{h} A+1 \) and the channel radius is \( a_c \).

Using the notation of Mughabghab [55] the strength function \( \tilde{S}_l \) is given by Eq. 2.17, where it is proportional to the sum of the reduced neutron widths.

\[
\tilde{S}_l = \frac{1}{(2l + 1)\Delta E} \sum_{\lambda} g_\lambda \Gamma^l_{\lambda,n} \tag{2.17}
\]

The reduced neutron widths are described by Eq. 2.18, where \( V_l \) is a pseudo penetrability.

\[
\Gamma^l_{\lambda,n} = \sqrt{\frac{1[eV]}{E_\lambda} \frac{\Gamma_{\lambda,n}}{V_l}} \tag{2.18}
\]

\( V_l \) is given in Eq. 2.19 where \( P_l \) is the hard sphere penetrability at \( E_\lambda \), and \( \rho = k_c a_c \) at \( E_\lambda \).

\[
V_l = \frac{P_l}{\rho} \tag{2.19}
\]
With these definitions, the contribution to the R-matrix from external levels can be determined from the known resonance parameters and used as prior input for SAMMY 8.1.

### 2.2 Resonance Self-Shielding

The RRR model requires parameters for each resonance and is limited by how well known those parameters may be. The limit of the energies at which the RRR model can be applied largely depends on the energy resolution of the highest resolution measurement of a given isotope. Often the measurements with the highest energy resolution are TOF measurements. There is uncertainty in the neutron energy inherent in all TOF measurements including, but not limited to: the time at which a neutron is emitted, detected, and the exact flight path a neutron traveled. In the example of TOF total cross section measurements this uncertainty has the effect of broadening the observed transmission, which can effectively average out some of the fluctuations in the true cross section. This is the reason why a URR model is necessary, because resonance fluctuations occur in the true un-observed cross section $\sigma_t(E)$ but cannot be observed due to the uncertainty in experiments and resonance overlap. The beginning of the URR for a measurement is often defined as where the uncertainty in TOF or $E$ is greater than the spacing of the resonances. If the observable is transmission, the effective average transmission $\langle T \rangle$ can be described as shown in Eq. 2.20, similar to the analysis by Semler [56].

$$\langle T(E) \rangle = \frac{\int_{E_1}^{E_2} \phi(E)e^{-\sigma_t(E)}dE}{\int_{E_1}^{E_2} \phi(E)dE} \quad (2.20)$$

Here the transmission is measured over some finite time bin $t_i$ with bin edges occurring at time $t_1$ and $t_2$ which corresponds to the energy bin from $E_1$ to $E_2$; $n$ is the areal density of the measured sample. In this case the uncertainty in neutron energy $\Delta E = E_2 - E_1$. We shall assume that the flux $\phi$ is constant in the energy range $E_1$ to $E_2$, and that the flux terms in the numerator and denominator cancel. After some manipulation we can define the $\langle T \rangle$ in terms of the average cross section $\langle \sigma_t \rangle$ in the bin $E_1$ to $E_2$ as shown in Eq. 2.21.

$$\langle T(E) \rangle = \frac{1}{E_2 - E_1} \int_{E_1}^{E_2} e^{-n[\sigma_t(E)-\langle \sigma_t \rangle + \langle \sigma_t \rangle]}dE \quad (2.21)$$
The pointwise transmission based on JEFF-3.3 cross section of Ta and $n=0.01$ is plotted with a solid green line. The average transmission taken over the TOF range $500 \rightarrow 2500 \ \mu s$ results in an $\langle T(\sigma_t) \rangle \approx 0.6$, which was plotted with the upper horizontal dashed green line. Taking the average of the cross section over the same energies, and calculating the transmission of the average cross section results in $T(\langle \sigma_t \rangle) \approx 0.2$, which is given by the lower red horizontal dashed line. This illustrates the importance of accounting for cross section fluctuation in measurements.

The average of the cross section in the energy bin $E_1$ to $E_2$ can be moved out of the integral as it is constant, and the final result is shown in Eq. 2.22 where there is a bracketed “fluctuation correction” term.

$$\langle T(E) \rangle = e^{-n(\sigma_t)} \left\{ \frac{1}{\Delta E} \int_{E_1}^{E_2} e^{-n[\sigma_t(E) - \langle \sigma_t \rangle]} dE \right\}$$

(2.22)

As all measurements are finite, the transmission is always measured over an average of some finite energy. Due to the non-linear relationship of transmission and cross section, in general, $T(\sigma_t(E)) \neq T(\langle \sigma_t(E) \rangle)$. If, however, the true un-observed cross section doesn’t fluctuate away from the average cross section over the energy between $E_1$ and $E_2$, the bracketed term in Eq. 2.22 is nearly unity and $\langle T(\sigma_t(E)) \rangle \approx T(\langle \sigma_t(E) \rangle)$. An extreme example of this inequality is illustrated in Fig 2.2.

In Fig. 2.2 the pointwise transmission plotted in solid green comes from the JEFF-3.3 cross section and $n = 0.01$. An average of this transmission was calculated over the TOF range $500 \rightarrow 2500 \ \mu s$ (209→8 eV) to be $\langle T(\sigma_t) \rangle \approx 0.6$, as illustrated by the green dashed line.
Figure 2.3: Transmission as a function of the total cross section $\sigma_t$ is plotted for two different sample thicknesses. The greater the sample thickness, the more non-linear the relationship between transmission and $\sigma_t$. The vertical dashed line represents the average cross section $\langle \sigma_t \rangle$. The JEFF-3.3 pointwise cross section was averaged over the same TOF, and the transmission of the average cross section was found to be $T(\langle \sigma_t \rangle) \approx 0.2$, as illustrated by the red dashed line. This inequality between $T(\langle \sigma_t \rangle)$ and $T(\langle \sigma_t \rangle)$ (popularly known as Jensen’s Inequality [57]) is due to the non-linear relationship between transmission and total cross section. This relationship is shown in Fig. 2.3 for samples of two different thicknesses. It is obvious from the plot that a thinner sample used in a transmission measurement results in a more linear relationship between transmission and $\sigma_t$. It can also be deduced from Eq. 2.22 that if the sample areal density $n$ (i.e. the sample thickness) is very small, the bracketed term can again be driven closer to unity. This leaves an experimentalist with two mechanisms to control the non-linearity of measured transmission and cross section: the resolution of the experiment (e.g., the bin width for which data are collected $\Delta tor\Delta E$), and the sample thickness.

This fluctuation effect is often called the “resonance self-shielding” effect, or perhaps more intuitively the “transmission enhancement” effect as the effective transmission is increased (effective cross section decreased) in thick self-shielded samples. To account for this fluctuation driven “resonance self-shielding” effect, parameters in the RRR are used to predict the fluctuating behavior in the URR. For the Monte Carlo code MCNP 6.1 [26]...
the probability table method is used. Probability tables contain cross section probability distributions at different energies; these are sampled during an MCNP calculation. The probability table method will be described further in §2.4. The MCNP URR model and its use of evaluated parameters for $^{181}$Ta are tested in this work with a thick-sample transmission measurement, where the sample is chosen to be very thick (large $n$) to highlight the self-shielding effect.

### 2.3 URR

As the incident neutron energy increases, the neutron widths increase proportionally to the square root of the energy $\Gamma_{\lambda n} = \gamma_{\lambda n}^2 2P_n \propto \sqrt{E}$ (for s-waves, here $P_{c=n}$ is the penetrability for a neutron channel). As a result, the nuclear levels naturally begin to overlap with each other. This, in addition to finite experimental resolution, leads to an energy region where resonances still affect the cross section but are no longer completely resolved, termed the URR. Accurately predicting average cross sections and variances in a region where resonances are being missed due to these reasons is accomplished by probabilistically calculating these quantities according to probability distributions for the average level spacing and average level widths. In 1956, Porter and Thomas found that a $\chi^2$ distribution was consistent with experimental resonance widths [58]. The distribution they described is now commonly referred to as the Porter-Thomas distribution with $\nu$ degrees of freedom. Typically this distribution is applied using one channel $c$ (and $\nu = 1$), but can be extended to multiple channels within a single reaction $x$ such as neutron widths for $I > 0$ and $l > 0$ or radiation widths. The Porter-Thomas distribution is shown in Eq. 2.23.

$$p(y)dy = \frac{\nu}{2} \frac{y^{\nu/2-1}e^{-\nu/2y}}{\Gamma(\frac{\nu}{2})}dy, \quad 0 < y \equiv \frac{\gamma_x^2}{\langle \gamma_x^2 \rangle} < \infty$$ (2.23)

$$\gamma_x^2 \equiv \sum_{c \in x} \gamma_c^2$$ (2.24)

Here the bold font $\Gamma$ represents the gamma function, not a nuclear level width. The term $\langle \gamma_x^2 \rangle = \nu \langle \gamma_c^2 \rangle$ where the $x$ is specific to a reaction and $c$ are the channels available for that reaction. Eq. 2.23 can be used for various partial widths, e.g.: reduced neutron widths, fission widths, and radiation widths. For reduced neutron widths, most often $\nu = 1$, but
the generalized Porter Thomas also applies to two-channel neutron widths $\nu = 2$, and for radiation widths $\nu$ is very large, which means that the distribution approaches a $\delta$ function.

In 1957 Wigner proposed a resonance spacing distribution that is now commonly referred to as Wigner’s Surmise [59]. Wigner surmised that resonances can be considered eigenvalues of a real symmetric matrix (and a complex Hermitian matrix), and that the levels would repel each other; specifically, that the probability for a single level spacing $S$ is proportional to $S$ itself. This causes neighboring spacings to be correlated, and the distribution was found to match experimental evidence very well. The distribution Wigner derived is given in Eq. 2.25 using the notation from Ref. [17] where $c \equiv \frac{\pi}{2D^2}$.

\[
p(S|D) = \left\{ e^{-c} \int_0^S S^dS' cS \right\} dS = \left\{ ce^{-cS^2/2S} \right\} dS \tag{2.25}
\]

Attempts have been made to improve the Wigner distribution many times, one of the most successful improvements was the Gaussian Orthogonal Ensemble (GOE) of nuclear Hamiltonian matrices $H$. This is presented in Eqs. 2.26 and 2.27 using the same notation as Ref. [17] and only briefly mentioned here for completeness as it wasn’t employed in the SAMMY 8.1 program. The value $N$ is the rank of matrix $H$ which is equal to the number of eigenvalues. $d(H) = \prod_{\mu<\nu} dH_{\mu\nu}$, and $\sigma$ is the spread of the eigenvalue spectrum. It is shown in Ref. [17] that the level spacing distribution as calculated by the GOE is nearly identical to the Wigner distribution.

\[
p(H|\sigma^2) = \prod_{\mu} e^{-\lambda H_{\mu\mu}^2} dH_{\mu\mu} \prod_{\mu<\nu} e^{-2\lambda H_{\mu\nu}^2} dH_{\mu\nu} \tag{2.26}
\]

\[
\lambda = \frac{N + 1}{4\sigma^2} \tag{2.27}
\]

SAMY 8.1 was used to evaluate URR cross sections for this study, therefore the equations described in the SAMMY manual [51] are presented here to define what formalism is being used to calculate the average cross sections. The total cross section is given by Eq. 2.28.

\[
\langle \sigma_c \rangle = \frac{2\pi g_c}{k_c^2} (1 - Re\langle S_{cc} \rangle) \tag{2.28}
\]
In Eq. 2.28 the average scattering matrix $\langle S_{cc} \rangle$ and the average $\mathfrak{R}$-matrix are defined as that in Eq. 2.29.

$$\langle S_{cc} \rangle = e^{-2i\phi_c} \frac{1 - \langle R_{cc} \rangle L_0^*}{1 - \langle R_{cc} \rangle L_0}, \quad \langle R_{cc} \rangle = R_c^\infty + i\pi s_c$$

(2.29)

$R_c^\infty$ is the distant level parameter and $s_c$ is the pole strength, which was defined in Eq. 2.14 and has been re-arranged to match the SAMMY formalism in Eq. 2.30. $L_0^*$ is the conjugate of $L_c$ which has been defined in §2.1 during the discussion of the $\mathfrak{R}$-matrix equations. The strength function $\tilde{S}$, shown in Eq. 2.30, is a function of reduced neutron widths $\Gamma_{n\lambda}^l$ at resonance energies $E_\lambda$ and has been described in Eq. 2.17 in §2.1.

$$s_c = \frac{\tilde{S}\sqrt{E}}{2\rho}, \quad \rho = k_c a_c$$

(2.30)

In Eq. 2.30 $k_c$ is a function of the incoming neutron energy $E$. The elastic scattering cross section is calculated as the difference in total cross section and non-elastic cross sections. Non-elastic cross sections are calculated using Hauser-Feshbach Theory with width fluctuations, given in Eq. 2.31.

$$\langle \sigma_{ab} \rangle = \frac{\pi g_a T_a T_b}{k_a^2 T} \int_0^\infty dt \ e^{-iT/\nu} \prod_{c \notin \gamma} \left( 1 + \frac{2T_c}{\nu_c} t \right)^{-\nu_c/2 - \delta_{ac} - \delta_{bc}}$$

(2.31)

The integrand in this expression is the width fluctuation correction factor, $a$ is the incident channel, $b$ is the exit channel, $T = \sum_c T_c$, where $T_c$ is the transmission coefficient for channel $c$, and $\nu_c$ is the number of degrees of freedom for that channel. The width fluctuation correction factor is according to the Moldauer prescription [60], and accounts for the fluctuation in the nuclear reaction widths that determine the cross section. $T_c$ for neutron channels is given in Eq. 2.32.

$$T_c = 1 - |\langle S_{cc} \rangle|^2 = \frac{4\pi P_c s_c}{|1 - \langle R_{cc} \rangle L_c|^2}$$

(2.32)

The photon and fission transmission coefficients are calculated with average parameters $\langle \Gamma_\gamma \rangle$, $\langle \Gamma_f \rangle$, and spin dependent mean level spacing $D_J$ as shown in Eq. 2.33. Calculation of $D_J$ is discussed in more detail in the SAMMY manual. The general method SAMMY employs to determine level spacing $D_J$ takes the user input level spacing of the s-wave levels close to
the neutron binding energy $D_{l=0}(E = 0)$ to determine the $J$-dependence, and the Gilbert-Cameron composite formula [61] to determine the energy dependence of the level spacing.

$$T_\gamma = 2\pi \frac{\langle \Gamma_\gamma \rangle}{D_J}, \quad T_f = 2\pi \frac{\langle \Gamma_f \rangle}{D_J} \quad (2.33)$$

### 2.4 Programming for the URR

To properly calculate neutron transport for the URR in computer programs, resonance self-shielding must be accounted for. As discussed in §2.2 the Monte Carlo program MCNP uses probability tables, and samples the probability distribution of the cross section at each neutron energy. Probability tables as used in MCNP 6.1 are created by the code NJOY21 [62]. NJOY21 is a nuclear data processing code designed to read ENDF/B format files and recreate cross section. In the URR NJOY21 uses the average resonance parameters listed in ENDF/B File 2 and randomly samples the Wigner and Porter Thomas probability distributions for the level widths and level spacing shown in Eqs. 2.23 and 2.25 to determine a random resonance ladder. Many random resonance ladders are generated to determine probability distributions of the cross section as a function of energy. Finally, these probability distributions are listed in “A Compact ENDF” (ACE) format in tables that can be efficiently sampled by the MCNP code.

SESH is a Monte Carlo program by Fröhner [63] which also predicts resonance self-shielding in the URR. SESH samples the Wigner and Porter Thomas distributions to calculate cross section and simulates neutron transport for simple geometries. SESH is designed to estimate the difference between theoretical cross section and the cross section that would be expected from a simulated observable such as transmission. Similar to NJOY21, SESH randomly samples from the Wigner and Porter Thomas distributions to generate “resonance pair” environments surrounding the user input average energies. A neutron’s energy is randomly placed between two resonances with spacing as sampled by the Wigner distribution, the partial widths of the two resonances are sampled from the Porter Thomas distribution, and the Doppler broadened cross section (using the Breit Wigner formula) is determined for resonances of that $J^\pi$. This is repeated for all spin sequences $J^\pi$ and the contribution of all spin sequences are summed. Multiple pairs of resonances are typically defined, each

---

1A sample Fortran code is given in Appendix A which demonstrates sampling from a simple Wigner distribution similar to the Fröhner calculation in SESH.
pair adding one resonance to the left and right of the neutron energy, creating a ladder of neutron resonances contributing to the cross section at that energy.

A typical use of the SESH program is to calculate the resonance self-shielding and multiple scattering yield corrections for transmission and capture yield measurements. SESH calculates the cross section as given by the input average resonance parameters, simulates a measurement (such as neutron transmission, capture yield, or self-indication) for a simple sample geometry, and calculates the ratio of the expected values $\sigma_\gamma$ and $\sigma_t$ to the values as determined by the simulated measurement $\sigma_{\gamma,\text{sim}}$ and $\sigma_{t,\text{sim}}$. These corrections have been used to correct the measured data in this work, which has been described further in §5.2. In §5.2 it is discussed how SESH was used iteratively with the fitting program FITACS, where SESH corrects data based on average resonance parameters and FITACS fits for updated average resonance parameters.
CHAPTER 3
MEASUREMENTS

Measurements for this work included datasets with two separate purposes: data which could be used to perform an evaluation, and data which could be used to validate evaluations. The measurements that were used to perform an evaluation were taken simultaneously at the RPI Linac with identical electron beam and target conditions; this is beneficial as having both the total and capture cross section measured in the same conditions allows for a more accurate evaluation of each quantity. The evaluation data set was measured at the RPI Linac during an entirely separate experiment from the validation experiment with different beam conditions, neutron target, etc. The goal of the evaluation dataset was to measure high resolution data that were not publicly available to the nuclear data community. The goal of the validation dataset was to stress test the current evaluated libraries for $^{181}$Ta (as well as a new RPI evaluation) in the RRR and URR in order to quantify which evaluation performs best. These two experiments have been summarized in Table 3.1 and elaborated upon in the following sections of this chapter. Items of interest in the evaluations include the extension of the RRR in the JEFF-3.3/JENDL-4.0 libraries, and the cross section fluctuation correction employed in the URR of ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0.

Table 3.1: Summary of measurements performed. The discussion of the details listed here is relegated to the proceeding sections of this chapter.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Evaluation</th>
<th>Validation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Overlap Filter</td>
<td>Enriched $^{10}$B</td>
<td>-</td>
</tr>
<tr>
<td>Neutron Producing Target</td>
<td>“C-shaped” Target</td>
<td>-</td>
</tr>
<tr>
<td>Electron Pulse Width [µs]</td>
<td>0.008</td>
<td>0.010</td>
</tr>
<tr>
<td>Beam Energy [MeV]</td>
<td>52</td>
<td>50</td>
</tr>
<tr>
<td>Pulse Repetition Rate [Hz]</td>
<td>400</td>
<td>400</td>
</tr>
<tr>
<td>Detector</td>
<td>MELINDA[64]</td>
<td>C$_6$D$_6$[50]</td>
</tr>
<tr>
<td>Energy Region [eV]</td>
<td>150-1.10$^5$</td>
<td>150-2·10$^5$</td>
</tr>
<tr>
<td>Flight Path Length [m]</td>
<td>100.141±0.01</td>
<td>45.27±0.05</td>
</tr>
<tr>
<td>Sample Thicknesses [at/b]</td>
<td>0.00566±1.4%</td>
<td>0.005631±0.8%</td>
</tr>
<tr>
<td></td>
<td>0.017131±0.7%</td>
<td>0.011171±1%</td>
</tr>
<tr>
<td></td>
<td>0.03358±0.3%</td>
<td>0.06716±0.4%</td>
</tr>
</tbody>
</table>
3.1 Evaluation Data

The evaluation data sets include: a transmission measurement at \(\approx100\) m, and a capture measurement at \(\approx45\) m. The 100 m transmission detection system has been described in detail in Ref. [64], and the 45 m capture system has been described in detail in Ref. [50]. Abbreviated descriptions of each system are provided here to improve the comprehension of the reader as well as to provide context for experimental considerations which were important to the work in this thesis. Each of these measurement systems were operated simultaneously on separate beam lines (100 m transmission on the center beam, 45 m capture on the East beam) at the RPI Linac. The RPI Linac produced electron bursts at a repetition rate of 400 Hz and accelerated them to approximately 50 MeV. These bursts of electrons were directed towards the “C-shaped” neutron producing target of Ta which has been described in detail in Ref. [66], but in brief is a set of Ta plates at the intersection of the electron beam axis and neutron beam axis. These Ta plates are cooled by circulating light water contained in a C-shaped aluminum can. The burst width of the electron pulses was approximately 8 ns. To reduce the amount of neutrons scattering off of air before entering the evacuated flight path, a large He tank approximately 4.5 m long was placed between the neutron target and the evacuated flight path.

3.1.1 100 m Transmission

Three samples of 1, 3, and 6 mm (0.00566, 0.017131, and 0.03358 at/b) thickness were measured for the transmission portion of the experiment. The samples are 99.95% pure elemental Ta, all derived from the same lot from the vendor. The sample dimensions and impurities are given in Appendix B. The RPI Linac has flight stations at approximately 15, 30, 100, and 250 m that could be used for a transmission experiment. The flight path (FP) of 100 m was chosen to optimize the count rate and energy resolution of the measurement. \(^{181}\)Ta has a reported average level spacing of 4.17 eV [55], which dictates the necessary resolution. The chosen experimental neutron burst width and flight path allowed for an energy resolution \(<4\) eV for the RRR energy range of interest (\(\approx 1 \rightarrow 4000\) eV), but neutron target thickness, and detector thickness negatively affect this resolution. The sample thicknesses and FP are important to satisfy the goals of the measurement: to record high resolution data up to approximately 4 keV, and statistically accurate data up to 100 keV.
3.1.1.1 Beam Characteristics

To reduce the probability for “overlap” neutrons (low energy neutrons which require longer than the 1/400 Hz = 2500 $\mu s$ to travel the FP) a 0.794 cm thick compressed powder disc of enriched $^{10}$B was placed in the center beam-line for the duration of the experiment. A 1.5875 cm (5/8 in) thick brick of Pb was mounted approximately 4.5 m from the neutron target in the beam path to reduce the number of Bremsstrahlung photons traveling from the target to the transmission detector. A 0.0254 cm (0.01 in) thick sheet of Co was placed in the beam for the duration of the experiment as part of the background determination method. The neutron beam was collimated before and after the sample position by brass inserts which had an inner diameter of 4.7625 cm (1.875 in).

3.1.1.2 Neutron Transmission Detector

The Mid-Energy LI-glass Neutron Detector Array (MELINDA) used $^6$Li doped glass scintillators placed in thin Al cases as neutron detectors. The detector consists of four modules, each module has a glass scintillator and two 12.7 cm (5 in) photomultiplier tubes (PMT) coupled to the Al case housing viewing the scintillating glass. The voltages and discriminator settings for the PMTs are listed in Table 3.2. The voltages listed in Table 3.2 come from gain-matching each of the multi-channel analyzer (MCA) spectra to the response of the weakest tube (Tube 7) for neutrons from a PuBe source, similar to Ref [64]. The MCA spectrum for each module can be seen in Fig. 3.1, where the gated and un-gated counts are shown for each detector based on whether the LLD and ULD have been applied.

Table 3.2: The voltages, lower level discriminator (LLD), and upper level discriminator (ULD) settings for the MELINDA system. The signal of the PMTs for each module is summed and passed through the same discriminator logic.

<table>
<thead>
<tr>
<th>Module</th>
<th>PMT 1</th>
<th>Voltage [V]</th>
<th>LLD [V]</th>
<th>ULD [V]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>-1160</td>
<td>-4.98</td>
<td>-10.0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>-1185</td>
<td>-4.98</td>
<td>-10.0</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>-1045</td>
<td>-4.18</td>
<td>-10.0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>-1215</td>
<td>-4.18</td>
<td>-10.0</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>-1110</td>
<td>-4.15</td>
<td>-10.0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>-1110</td>
<td>-4.15</td>
<td>-10.0</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>-1155</td>
<td>-3.63</td>
<td>-10.0</td>
<td></td>
</tr>
<tr>
<td></td>
<td>-1130</td>
<td>-3.63</td>
<td>-10.0</td>
<td></td>
</tr>
</tbody>
</table>
Figure 3.1: The gated and un-gated MCA spectra are shown with the red and blue lines, respectively. The gated counts come from the voltage pulses in the PMTs which pass the LLD and ULD. The MCA was collected using a 1024 bin structure.

The discriminator filtered signal from all four modules is summed and the detector interaction time $t_i$ is recorded as a function of TOF. A Comtec MCS6A Multi-stop TDC TOF clock was set to record events within a 0.8 ns bin width $b_w$ up to TOF=2500 $\mu$s. An image of the MELINDA system is shown in Fig. 3.2, where the four modules are aligned to the center of the neutron beam, approximately 1.7 m from the floor.

In the process of matching the gain of the detectors, a new reference source setup was designed. This new design is meant to moderate the neutrons from the source, shield the detector from gammas emitted from the source, and reduce the dose received by the researcher performing the gain alignment. The source was placed into a 10.16 cm (4 in) polyethylene block to moderate the neutrons, and two 5.08 cm (2 in) bricks of Pb are placed between the polyethylene moderator and the detector. This geometry is shown in Fig. 3.3. In a crude measurement using a Geiger counter, the estimated reduction in dose to a researcher using this new geometry is 2-3 times less than the previous geometry. The average count rate for this geometry with the source in the reference position is approximately 2000 cps.
Figure 3.2: The Mid Energy LI-glass Neutron Detector Array (MELINDA).
Each of the four aluminum cases hold $^6$Li doped scintillating glass, with 2 PMTs coupled to the light-tight aluminum module. The detector resides in the RPI Linac 100 m detector station.

The source position in relation to the detector can be seen in Fig. 3.4. The source is kept on the end of a 3.5 m Al rod, which is seen in the images to the left and right of Fig. 3.4. The source position in the new reference geometry is marked by a red circle in the images to the left and right, each image showing a different angle of the new position in reference to the MELINDA detector.

3.1.2 45 m Capture

Two samples of 1 and 2 mm thicknesses are measured for the capture portion of the experiment. The 1 mm (0.005631 at/b) sample is 99.95% pure elemental Ta (see Appendix B) and comes from the same batch as the samples used in the transmission measurement, the 2 mm (0.011150 at/b) sample is described in Ref. [50]. Along with the samples of interest, a sample of B$_4$C with 91.7% enriched $^{10}$B [67] and 1.30 cm thick was used to measure the neutron rate reaching the sample, and a 1 cm thick natural Pb sample was used to measure
Figure 3.3: An illustration of the new reference source position for the 100 m transmission detector MELINDA. This geometry was used to perform a reference count rate for the MELINDA detector before each measurement. Starting from the right: the yellow rectangle represents the 10.16 cm (4 in) polyethylene block with a bore hole that houses the source, followed by a gray block that represents two 5.08 cm (2 in) Pb bricks.

the in-beam photon scattering background (see §4.2.1 and Ref. [50]). Neutron capture systems already implemented at the RPI Linac are the 16 segment NaI multiplicity detector [68] at a FP of 25 m, and the C₆D₆ low mass capture detector [33] at 45 m. As the NaI detector is mostly limited to neutron energies below a few keV, the C₆D₆ capture system was best suited for the goals of this measurement: resolving resonances up to approximately 4 keV, and measuring statistically accurate cross section up to 100 keV.

3.1.2.1 Beam Characteristics

Simultaneously with the 100 m transmission experiment on the center beam line, the capture system is operated along the East beam line. To remove Bremsstrahlung photons in the beam originating from the electron interaction in the neutron target, 1.5875 cm (5/8 in) of Pb was placed in the beam at approximately 15 m. To remove the “overlap” neutrons, which have been described in §3.1.1.1, a 0.396875 cm (5/32 in) thick B₄C plate was placed in the neutron beam. A 4.7625 cm (1.875 in) inner diameter brass collimator is placed at approximately 20 and 21 m. Downstream of the brass collimators are polyethylene and Pb collimators each of which has a 5.08 cm (2 in) inner diameter. Both the Pb
brick and polyethylene collimators were 5.08 cm (2 in) thick and located at a flight path of approximately 35 m from the neutron target (same geometry as Ref. [50]).

3.1.2.2 Neutron Capture Yield Detector

The capture detector is an aluminum structure that suspends a sample of interest and four C₆D₆ photon detectors. The system was designed to reduce the probability for neutron capture by anything other than the sample of interest. For this reason all of the structural materials are aluminum, as well as the housing for the C₆D₆ detectors and PMTs. An image of the capture system is shown in Fig. 3.5.
Figure 3.5: The capture detector system at a flight path of approximately 45 m. Four C$_6$D$_6$ detectors are suspended by aluminum rods and face the sample of interest. Sample material was changed by an automated step motor, which moved the sample holders to the left and right (orthogonal to the neutron beam).

The detector with the largest expected gain was tested with a $^{22}$Na source to ensure that the signal voltage did not exceed the dynamic range of the recording system. Once the voltage on the detector with the largest gain was set, all the other detectors were aligned to it using $^{22}$Na and $^{137}$Cs gamma sources. The voltages applied to the PMTs during the Aug. 2017 Ta measurement are listed in Table 3.3.

Table 3.3: The voltages for the PMTs in the 45 m capture detection system during the Aug 2017 Ta measurement.

<table>
<thead>
<tr>
<th>PMT</th>
<th>Voltage [V]</th>
</tr>
</thead>
<tbody>
<tr>
<td>PMT 1</td>
<td>-1801</td>
</tr>
<tr>
<td>PMT 2</td>
<td>-1840</td>
</tr>
<tr>
<td>PMT 3</td>
<td>-1804</td>
</tr>
<tr>
<td>PMT 4</td>
<td>-1634</td>
</tr>
</tbody>
</table>

This process of alignment with the gamma sources is also used to determine an energy calibration for the capture detectors. An MCA of pulse integrals is taken while the detector
is viewing a gamma source and the Compton edge [69] can be observed for each characteristic decay. The $^{22}\text{Na}$ source has characteristic decay lines at 1.275 MeV and 0.511 MeV (annihilation photons) and $^{137}\text{Cs}$ has a decay line at 0.662 MeV. The pulse integral measured at each Compton edge corresponding to the characteristic decays has been plotted in Fig. 3.6 for each detector. Along with the discrete points, a fitted line for each detector is shown. The equation $PI(E_c) = a \cdot E_c + b$ is fitted for parameters $a$ and $b$, where $PI$ is the pulse integral and $E_c$ is the Compton edge energy. The Compton edge energy can be found by Eq. 3.1 [69] where $E_\gamma$ is the photon energy and the units are keV. These fitted parameters are used to determine the energy deposited in the detector during the experiment.

![Figure 3.6: The measured pulse integral for each discrete Compton edge energy is plotted along with fitted lines for each detector. The energy of the decay gammas are given in parentheses. The parameters from the fit are used in post-processing to determine the energy deposited in the detectors.](image)

$$E_c(E_\gamma) = \frac{2.0E_\gamma^2}{511.0 + 2.0E_\gamma} \tag{3.1}$$

During the TOF data measurement the signals from each of the four detectors was digitized and recorded as a function of time with an SIS3305 10-bit digitizer with a sample rate of 1.25 GS/s. In this way, all of the information contained in each voltage pulse was
saved and written to file. The advantage of the digitized data (as opposed to simply recording that an event occurred at time $t_i$, as was done for the transmission detector) is the freedom to process the voltage pulses in many different ways. For example, the LLD and ULD are fixed quantities in the transmission detector system and any event that does not pass the LLD/ULD logic will never be recorded, but in the capture system the LLD can be chosen to optimize the experiment in the post-processing.

3.2 Validation Data: Thick Sample Transmission

The validation data contains a single dataset: the transmission for a 12 mm thick Ta sample. This measurement was made at 35 m with an in-beam $^6$Li doped scintillator viewed by two PMTs along the East beam line; this detector is fully described in Ref. [65] and can be seen in Fig. 3.7. The neutron production target used was the Bare Bounce Target (BBT) [66]: an array of Ta moderated by water to the side of the neutron beam axis, and an on-axis 2.54 cm (1 in) thick polyethylene disc to moderate and reflect neutrons along the neutron beam axis. This geometry is shown in Fig. 1.14. The Linac was operated at a repetition rate of 400 Hz, accelerating electrons to approximately 50 MeV. The burst width of the electron pulses was approximately 10 ns.
Figure 3.7: The 35 m $^6$Li doped glass scintillator detection system. The detector resides along the East beam line at the RPI Linac, as indicated by the label on the bottom window sill.

The flight path of 35 m was chosen to improve the count rate as opposed to the 100 m transmission detector, as well as maintain better resolution than a transmission measurement at 15 m (these being the most practical flight paths at the RPI Linac). The advantage gained from using the BBT as opposed to the “C-shaped” target was a reduction of in-beam Bremsstrahlung photons, which in turn reduces the time dependent background in the measurement. This reduction in photon background is due to the combination of the neutron production target being off-axis, and the inclusion of a 20.32 cm thick Pb slab shadow-shield attenuating Bremsstrahlung photons (see Fig. 1.14). It should be noted that there was a trade-off between using the “C-shaped” target and the BBT. The BBT has a reduced photon background component but produces a neutron beam of lower intensity in the energy region of interest. The signal-to-background ratio (on the East beam, with 35 m flight path) of the
“C-shaped” target is 5:1 for most of the energy region of interest, and between 10:1 and 90:1 for the BBT.

Two stacked 6 mm thick 99.95% pure Ta samples served as the 12 mm thick (0.06716 at/b) Ta sample measured for the validation dataset, these samples are described further in Appendix B and can be seen in Fig. 3.8. This thickness served to highlight the resonance self-shielding effect found in the URR (see §2.2). A 0.0821 at/b depleted U sample was measured simultaneously with the 12 mm thick Ta sample to verify experimental parameters such as the FP, $t_0$, and estimated experimental background.

![Figure 3.8: Two 6 mm thick Ta samples are stacked to form the 12 mm thick sample used in the thick-sample validation transmission measurement.](image)

### 3.2.1 Beam Characteristics

A 0.396875 cm (5/32 in) thick $B_4C$ plate was kept in the beam for the duration of the experiment to reduce the probability for “overlap” neutrons. As part of the background determination method a 0.0254 cm thick (0.01 in) sheet of Co and a 2.54 cm (1 in) thick plate of Al was kept in the beam at approximately 15 m for the duration of the experiment. To remove in-beam Bremsstrahlung photons and Ta decay photons, a 1.27 cm (0.5 in) thick plate of Pb was placed in the beam at approximately 15 m. The sample changer resided at a FP of approximately 20 m. Brass inserts with a 4.7625 cm (1.875 in) inner diameter were placed in the beam before and after the sample to collimate the beam.
3.2.2 Neutron Transmission Detector

The detector residing at 35 m is an in-beam $^6\text{Li}$ doped glass scintillator, housed in an Al case. Two PMTs which are coupled to the Al housing view the scintillating glass and detect scintillation light. The PMTs were gain matched using the PuBe neutron source in the reference position. The reference position for this experiment is shown in Fig. 3.9.

![Diagram showing the reference source geometry for the 35 m transmission detector.](image)

This reference count rate is measured to ensure that the detector is behaving normally before collecting the TOF data of interest. The direct distance from the source to the center of the glass scintillator is approximately 2.6 m. (Note: not to scale.)

In Fig. 3.9 it is illustrated that the reference PuBe neutron source was moderated by a 5.08 cm thick polyethylene brick, and the decay photons are attenuated by the 5.08 cm thick Pb brick. The reference source is used to align the responses of the PMTs and verify that the LLD and ULD have been effectively chosen. The signal of the two PMTs was summed and gated by the same LLD and ULD. The voltages and discriminator settings are given in Table 3.4.
Table 3.4: The voltages for the PMTs in the 35 m transmission detector system, along with the associated discriminator settings for the system. The signal from both PMTs is summed and passed through the same discriminator logic.

<table>
<thead>
<tr>
<th></th>
<th>Voltage [V]</th>
<th>LLD [mV]</th>
<th>ULD [mV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>PMT 1</td>
<td>-1064</td>
<td>-0.58</td>
<td>-1.36</td>
</tr>
<tr>
<td>PMT 2</td>
<td>-1149</td>
<td>-0.58</td>
<td>-1.36</td>
</tr>
</tbody>
</table>
CHAPTER 4
DATA REDUCTION

4.1 Neutron Transmission

Transmission is the ratio of count rates for when a sample is in and out of the neutron beam. This is shown in Eq. 4.1, where the “sample in” count rate is defined as \( \dot{C}_{Ta}(t_i) \), and the “sample out” is defined as \( \dot{C}_o(t_i) \). The neutron detector being used to measure these count rates resided in the beam after the sample location, as a result of this there was a strong TOF-dependent background included in the count rates. The background changes with the beam configuration such as “sample in” and “sample out”. Each time-dependent background is assumed to have the same shape \( \dot{B}(t_i) \). This shape was then normalized by the coefficients \( k_{Ta}, k_o \) determined by the count rate at the fixed notch resonance (discussed further in §4.1.2) for the “sample in” and “sample out” configurations, respectively. The respective constant backgrounds, as indicated by the subscript are defined as \( \dot{B}_{0Ta} \) and \( \dot{B}_{0o} \). The values of \( \alpha_{1,2,3,4} \) represent the cycle by cycle monitor normalization. They are typically close to unity, but have associated uncertainty (see Appendix F) which is propagated to the final correlation matrix. For the sake of brevity, the time dependence \( t_i \) is replaced by the subscript \( i \).

\[
T_i = \frac{\alpha_1 \dot{C}_{Ta,i} - \alpha_2 k_{Ta} \dot{B}_i - \dot{B}_{0Ta}}{\alpha_3 \dot{C}_{o,i} - \alpha_4 k_o \dot{B}_i - \dot{B}_{0o}}
\]  

(4.1)

The counts vectors have been converted to count rates by division of the time that a given channel \( t_i \) in TOF is collecting counts data; this is shown in Eq. 4.2. The quantity \( b_w \) is the width in time a channel \( t_i \) is open, and \( trig \) is the number of Linac triggers which corresponds to the number of times the bin \( t_i \) was open to collect data. \( C(t_i) \) are the dead-time corrected counts (see §4.1.1) for each time bin \( t_i \).

\[
\dot{C}(t_i) = \frac{C(t_i)}{b_w trig}
\]

(4.2)

The experimental transmission is used to calculate the microscopic neutron total cross section \( \sigma_t \). This relation is shown in Eq. 4.3, where the sample thickness \( n \) has units of \([\text{barn}^{-1}]\).
\[ T = e^{-n\sigma t} \quad (4.3) \]

### 4.1.1 Dead Time

Dead time is defined by Knoll [69] as the “minimum amount of time which must separate two events in order that they be recorded as two separate pulses”. This is determined by the detector being used as well as the set of electronics monitoring the detector. To obtain the true count rate the observed count rate must be corrected by the dead time \( \tau \) of the system. The dead time equation is the same for the transmission and capture yield experiments presented in this work, and assumed to follow closely to the nonparalyzable dead time model. The dead time correction employed in this work is derived from the work of Danon [70]. There are two different scenarios: dead time which is less than the raw bin width \( \tilde{b}_w \) of the detection system \( \tau < \tilde{b}_w \), and dead time which is greater than the raw bin width of the system \( \tau > \tilde{b}_w \). If the dead time is less than the raw bin width of the system, the familiar dead time correction as described by Knoll [69] can be applied to the count rate; the equation for this type dead time correction factor \( dtcf \) is given in Eq. 4.4. The term \( trig \) is the integer number of times that the Linac was pulsed, and number of times each time bin \( t_i \) was open collecting data. \( C_i = C(t_i) \) as described previously.

\[
dtcf_{\tau < \tilde{b}_w,i} = \frac{1}{1 - \frac{C_i \tau}{trig \tilde{b}_w}} \quad (4.4)
\]

If, however, the dead time was greater than the raw bin width of the data collection system, which was the case for the capture and transmission systems used in this work, the dead time correction must be taken as shown in Eq. 4.5.

\[
dtcf_{\tau > \tilde{b}_w,i} = \frac{1}{1 - \sum_j C_j w_j} \quad (4.5)
\]

The weights \( w_j \) in Eq. 4.5 are determined in an algorithmic fashion for each bin covered by the dead time, where all weights are 1.0 except the first and last bin. An example of the dead time correction is illustrated in Fig. 4.1. In the illustrated example, the dead time covers 3.1 bin widths. The counts in each bin \( C_i \) at time \( t_i \) are corrected by \( j \) bins that precede the bin at \( t_i \). In this example, the counts in bin \( i \) of Fig. 4.1 are being corrected. As the true time at which an event occurs in bin \( t_i \) is unknown, it was assumed that the detector was
dead until the average time in the bin (the midpoint of the bin). For this reason we weight the counts by \( w_{j=0} = 0.5 \) in the current bin \( i \). We use the counts occurring in the preceding \( j \) bins to correct \( C_i \) starting at the midpoint of the current bin. The weight of the last bin is the fraction of the bin which the dead time spans (starting at the midpoint of bin \( i \)), in this case \( w_{j=3} = 0.6 \).

\[
\begin{align*}
  w_j &= i - 3 & i - 2 & i - 1 & i \\
  0.6 & & 1.0 & 1.0 & 0.5
\end{align*}
\]

Figure 4.1: An illustration of the dead time correction employed for the count rates used for the transmission and capture yield measurements. The dead time is indicated by the red object occurring over several TOF bins. Each bin at time \( i \) is corrected by \( j \) many bins that the dead time spans preceding the midpoint of bin \( i \). Dead time is determined for each system, and applied in the same fashion for each.

4.1.2 Background

For the transmission measurements contained in this Ph.D. work, the background was determined by the “black resonance” method. The black resonance method is where a material with a strong isolated resonance will remove almost all on-energy neutrons close to the resonance energy creating a “notch” in the transmission. This method has been used widely and its strengths and weaknesses have been discussed in detail by Syme [71]. To determine the background, a portion of the experiment was dedicated to measuring the same “sample in” and “sample out” quantities of \( \dot{C}_{T,n} \) and \( \dot{C}_{o,n} \) but with extra notch material in the beam, focusing on the count rates at the notch resonance energies. The count rates at the notch energies are nearly the sum of the time dependent and constant background. The components of the background include constant room background \( \dot{B}_0 \), time dependent gamma background \( \dot{B}_{\gamma}(t_i) \), and time dependent off-energy neutron background \( \dot{B}_n(t_i) \) as described in Eq. 4.6.
\[
\dot{B}(t_i)_{\text{tot}} = \dot{B}_\gamma(t_i) + \dot{B}_n(t_i) + \dot{B}_0
\] (4.6)

For the measurements presented herein the neutron and photon backgrounds were summed and fitted together as \(\dot{B}(t_i) = \dot{B}_\gamma(t_i) + \dot{B}_n(t_i)\). When employing the black resonance background method there is often a “fixed notch” material, which is a notch material that resides in the beam for the duration of the experiment. This notch must be carefully selected to ensure that it does not remove neutrons in the energy region of interest. For example, in the thick-sample validation transmission measurement introduced in §3.2 there were two fixed notches, Co and Al which have strong resonances at 132 eV and 35 keV, respectively. These notches were chosen to minimize the potential cross section of the notch materials and constrain the background shape at both 132 eV and 35 keV. The fixed notches were used to determine \(k_{Ta}\) and \(k_o\) which normalized the beam shape that was fitted to the notch points obtained from the dedicated notch runs. When feasible, the method of using two fixed notches is preferrable to one fixed notch, as it can provide a concrete measurement of the background at two energies for all data sets. Using two fixed notches justifies that the single normalization coefficient is reasonably accurate at the energies between those notch points. An example of a notch resonance is presented in Fig. 4.2.
In Fig. 4.2 the Co resonance at 132 eV removed almost all the neutrons in the beam at that energy. The discrete background point for the Co notch is the mean value of the count rate shown in the delineated region. The propagated error of the mean was the uncertainty on that background point. The same process was used for all of the notches measured in both the 100 m and 35 m transmission measurements.

### 4.1.2.1 Background: 100 m Transmission

The notch materials used in this experiment were Mn, Na, Al, and Mg which have strong resonances at approximately 0.33, 2.85, 35.0, and 83 keV respectively. A background count rate was determined at each of these points and fitted with the function Eq. 4.7 which was subtracted off of the “sample in” and “sample out” count rates. This can be seen...
in Fig. 4.3. The constant backgrounds $\dot{B}_{0Ta}$, $\dot{B}_{0o}$ are the average count rates at the longest TOF in the spectrum for each sample configuration (approximately 2450 µs).

$$\dot{B}(t_i) = a \cdot t_i^{-b}$$ (4.7)

The background function was normalized to the Co fixed notch at 132 eV using the $k_{Ta}$ and $k_o$ normalization coefficients.

Figure 4.3: The count rates measured by the MELINDA detector at a flight path of 100 m. The background counts and fitted background functions for the “open” and “sample in” configurations are given by the blue and red circles and lines, respectively. The background functions are normalized to the fixed notch (132 eV resonance of Co) at an approximate TOF of 625 µs.

4.1.2.2 Background: 35 m Thick-Sample Transmission

Measurements to determine the background for the validation transmission measurement included the sample in and sample out count rates with the following “notches”: the 250 keV Li resonance, 35 keV Al resonance, 2.8 keV Na resonance, and 132 eV Co resonance. The average notch count rate for “sample in” and “sample out” measurements at the described resonance energies were the discrete count rates which were fitted. The background for the validation transmission measurement is modeled by the exponential function in Eq.
4.8. This method of background treatment introduces correlations between the transmissions calculated for different time bins.

\[ \dot{B}(t_i) = a \cdot e^{-bt_i} \]  \hspace{1cm} (4.8)

The average background count rate for the sample in and sample out configurations were then normalized to the fixed notch count rate at the 35 keV Al resonance with parameters \( k_{Ta} \) and \( k_o \), as seen in Eq. 4.1. The final count rates, backgrounds, and notch count rates are shown in Fig. 4.4.

![Figure 4.4: The count rate for the thick-sample transmission measurement. The upper solid green curve represents the measured open beam count rate and the dashed green curve matching the green circular points is the calculated open background count rate. The solid blue line represents the sample count rate and the dashed blue line matching the blue circular points is the calculated sample background count rate. Blue and green circles indicate “black resonance” count rates for the sample in and open count rates, respectively.](image-url)
4.1.3 Uncertainty and Correlation

The uncertainty $\Delta T$ found in the transmission (Eq. 4.1) is obtained from a combination of statistical uncertainty derived from Poisson counting statistics [69] for each of the measured count rates and systematic uncertainties from normalizations by monitors $\alpha_{1,2,3,4}$, $k_{T_a}$, $k_O$ and the parameters of the background functions $a$ and $b$. The uncertainty $\Delta T$ comes from the diagonal elements of matrix $C_y$, which is given by Eq. 4.9 where $C_x$ is the input covariance matrix and $F_x$ is the Jacobian matrix. The Jacobian matrix, in this case, consists of the derivatives of each bin of the transmission equation with respect to each variable in the transmission equation.

$$C_y = F_x C_x F_x^T$$  \hspace{1cm} (4.9)

In defining the input covariance $C_x$, we begin by assuming that each TOF channel of each count rate vector is independent of the other count rates and normalizations. We then form the covariance matrix $C_x$ that contains each of the constituents of the transmission equation for every TOF. This results in a matrix which is $m \times m$, where $m = (p_{stat} \cdot N + p_{sys})$ taking the variable $p_{stat}$ as the number of statistical (independent) parameters, $N$ as the number of energy or TOF points in the transmission, and $p_{sys}$ as the number of systematic parameters. An example is shown in Eq. 4.10.

$$C_x = \begin{bmatrix}
\Delta C_{s,i} & 0 & \ldots & \ldots & 0 \\
0 & \Delta C_{o,i} & \ldots & \ldots & \vdots \\
\vdots & \vdots & \ddots & \vdots & \vdots \\
\Delta C_{s,N} & \Delta C_{o,N} & \Delta k_{T_a} & \Delta k_O & \Delta a \Delta b \\
\Delta b \Delta a & \Delta b & \ldots & \ldots & \ldots \\
0 & \ldots & & & 
\end{bmatrix}$$ \hspace{1cm} (4.10)
Here the ellipses (...) indicate the continuation of zeros as well as the continuation of the time dependent variables for count rates and constant variables which could not be included due to space limitations. It should be noted that the variables with subscript $i$ increase by 1 and go to $N$ ($i \in 0, i+1, ..., N$). The covariance matrix of the input quantities $C_x$ is nearly diagonal based on the assumed independence of most of the variables. Starting with the initial notation as found in Ref. [72] the energy to energy covariance of the transmission $cov(T_i, T_j) = C_y = F_x C_x F_x^T$. The matrix $F_x$ is the sensitivity matrix of the transmission to each of it’s constituents given by the Jacobian, as shown in Eq. 4.11.

$$F^T = \begin{bmatrix} \frac{\partial T_i}{\partial C_{s,i}} & \cdots & \frac{\partial T_i}{\partial C_{s,N}} \\ \frac{\partial T_i}{\partial C_{o,i}} & \cdots & \frac{\partial T_i}{\partial C_{o,N}} \\ \vdots & \cdots & \vdots \\ \frac{\partial T_N}{\partial C_{s,i}} & \cdots & \frac{\partial T_N}{\partial C_{s,N}} \\ \frac{\partial T_N}{\partial C_{o,i}} & \cdots & \frac{\partial T_N}{\partial C_{o,N}} \\ \frac{\partial T_i}{\partial a} & \cdots & \frac{\partial T_i}{\partial a} \\ \frac{\partial T_i}{\partial b} & \cdots & \frac{\partial T_i}{\partial b} \\ \frac{\partial T_i}{\partial k_{Ta}} & \cdots & \frac{\partial T_i}{\partial k_{Ta}} \\ \frac{\partial T_i}{\partial k_o} & \cdots & \frac{\partial T_i}{\partial k_o} \\ \frac{\partial T_i}{\partial \alpha_1} & \cdots & \frac{\partial T_i}{\partial \alpha_1} \\ \frac{\partial T_i}{\partial \alpha_2} & \cdots & \frac{\partial T_i}{\partial \alpha_2} \\ \frac{\partial T_i}{\partial \alpha_3} & \cdots & \frac{\partial T_i}{\partial \alpha_3} \\ \frac{\partial T_i}{\partial \alpha_4} & \cdots & \frac{\partial T_i}{\partial \alpha_4} \\ \frac{\partial T_i}{\partial B_{0, Ta}} & \cdots & \frac{\partial T_i}{\partial B_{0, Ta}} \\ \frac{\partial T_i}{\partial B_{0, o}} & \cdots & \frac{\partial T_i}{\partial B_{0, o}} \end{bmatrix}$$

(4.11)

The sensitivity matrix is an $Nx m$ matrix. The horizontal ellipses indicate that the derivative of every transmission $T_i$ is taken at each point in the TOF spectrum $i \rightarrow N$. The vertical ellipses indicate that the derivative of each $T_i$ is taken with respect to the count rate at each point in the TOF spectrum $i \rightarrow N$. Using this notation would result in very large matrices which potentially require large amounts of memory. It should also be noted that many elements of these matrices would be filled with zeros. To reduce memory requirements, simplify the computation, and highlight the contribution of the correlations, the correlated (statistical) and un-correlated (systematic) parameters have been separated into two matrices.
similar to the method shown by Becker et. al. [73]. This is shown in Eq. 4.12. Each of the terms in the right-hand side of Eq. 4.12 has been expanded in Eqs. 4.13, 4.14 and 4.15.

\[
C_y = F_x C_x F_x^T = F_{st} C_{st} F_{st}^T + F_{sy} C_{sy} F_{sy}^T
\] (4.12)

\[
F_{st} C_{st} F_{st}^T = [F_{st}]
\]

\[
\begin{bmatrix}
\Delta C_{s,i}^2 & \cdots & 0 \\
\Delta C_{o,i}^2 & \ddots & \\
0 & \cdots & \Delta C_{s,N}^2 \\
\end{bmatrix}
= 
\begin{bmatrix}
\frac{\partial T_i}{\partial C_{s,i}} & \cdots & \frac{\partial T_N}{\partial C_{s,i}} \\
\frac{\partial T_i}{\partial C_{o,i}} & \ddots & \\
\frac{\partial T_i}{\partial C_{s,N}} & \cdots & \frac{\partial T_N}{\partial C_{o,N}}
\end{bmatrix}
\] (4.13)

\[
C_{sy} = 
\begin{bmatrix}
\Delta a^2 & \Delta b \Delta a & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\Delta a \Delta b & \Delta b^2 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & \Delta k_{Ta}^2 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & \Delta k_o^2 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & \Delta B_{0,Ta}^2 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & \Delta B_{0,o}^2 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & \Delta \alpha_1^2 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & \Delta \alpha_2^2 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \Delta \alpha_3^2 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \Delta \alpha_4^2
\end{bmatrix}
\] (4.14)
\[
\mathbf{f}_{sy}^T = \begin{bmatrix}
\frac{\partial T_i}{\partial a} & \cdots & \frac{\partial T_i}{\partial a} \\
\frac{\partial T_i}{\partial b} & \frac{\partial T_i}{\partial b} & \frac{\partial T_i}{\partial b} \\
\frac{\partial T_i}{\partial k_{Ta}} & \frac{\partial T_i}{\partial k_{Ta}} & \frac{\partial T_i}{\partial k_{Ta}} \\
\frac{\partial T_i}{\partial k_o} & \frac{\partial T_i}{\partial k_o} & \frac{\partial T_i}{\partial k_o} \\
\frac{\partial T_i}{\partial B_{0T_i}} & \frac{\partial T_i}{\partial B_{0T_i}} & \frac{\partial T_i}{\partial B_{0T_i}} \\
\frac{\partial T_i}{\partial B_{0i}} & \frac{\partial T_i}{\partial B_{0i}} & \frac{\partial T_i}{\partial B_{0i}} \\
\frac{\partial T_i}{\partial \alpha_1} & \frac{\partial T_i}{\partial \alpha_1} & \frac{\partial T_i}{\partial \alpha_1} \\
\frac{\partial T_i}{\partial \alpha_2} & \frac{\partial T_i}{\partial \alpha_2} & \frac{\partial T_i}{\partial \alpha_2} \\
\frac{\partial T_i}{\partial \alpha_3} & \frac{\partial T_i}{\partial \alpha_3} & \frac{\partial T_i}{\partial \alpha_3} \\
\frac{\partial T_i}{\partial \alpha_4} & \frac{\partial T_i}{\partial \alpha_4} & \frac{\partial T_i}{\partial \alpha_4}
\end{bmatrix}
\]  
(4.15)

As the derivatives \( \frac{\partial T_i}{\partial C_{s,i}} \) and \( \frac{\partial T_i}{\partial C_{o,i}} \) are only non-zero when \( T_i \) and \( C_{s,i}, C_{o,i} \) correspond to the same channel, Eq. 4.13 reduces to the diagonal \( N \times N \) matrix given in Eq. 4.16. The derivatives of transmission with respect to all the constituents of Eq. 4.1 are listed in Eq. 4.17. To simplify the equations, the numerator and denominator of Eq. 4.1 are denoted by \( N_i \) and \( D_i \) respectively.

\[
\begin{bmatrix}
\left( \frac{\partial T_i}{\partial C_{s,i}} \Delta C_{s,i} \right)^2 + \left( \frac{\partial T_i}{\partial C_{o,i}} \Delta C_{o,i} \right)^2 & \cdots & 0 \\
\cdots & \cdots & \cdots & \cdots \\
0 & \left( \frac{\partial T_N}{\partial C_{s,N}} \Delta C_{s,N} \right)^2 + \left( \frac{\partial T_N}{\partial C_{o,N}} \Delta C_{o,N} \right)^2
\end{bmatrix}
\]  
(4.16)

\[
\begin{align*}
\frac{\partial T_i}{\partial C_{s,i}} &= \frac{\alpha_1}{D_i} \\
\frac{\partial T_i}{\partial C_{o,i}} &= \frac{N_i \alpha_3}{D_i^2} \\
\frac{\partial T_i}{\partial k_{Ta}} &= -\frac{\alpha_2 B_{T_i}}{D_i} \\
\frac{\partial T_i}{\partial k_o} &= \frac{-N_i \alpha_4 B_{T_i}}{D_i^2} \\
\frac{\partial T_i}{\partial B_{0T_i}} &= \frac{\left( k_{T_0} \alpha_2 D_i + k_o \alpha_4 N_i \right) B_{iT_i}}{D_i^2} \\
\frac{\partial T_i}{\partial B_{0i}} &= \frac{\left( k_{T_0} \alpha_2 D_i + k_o \alpha_4 N_i \right) e^{-b_{T_i}}}{D_i^2}
\end{align*}
\]  
(4.17)

4.1.4 Reporting Data Covariance

When reporting experimental nuclear data covariance, often the entire matrix is not given. This is often due to space limitations, e.g., if a transmission dataset contains \( N \) energy points, then the energy-to-energy covariance requires \( N^2 \) numbers. The covariance matrix
Figure 4.5: An example file format for reporting experimental data covariance. The orange box indicates systematic uncertainties ($\Delta k_{Ta}, \Delta k_o$, etc.). The blue box indicates correlated input: a covariance matrix for some of the systematic variables ($\Delta a^2, \Delta a\Delta b$, etc.). The red box contains vectors in the order of: energy, observable ($T$), and uncertainty on observable ($\Delta T$). The green box indicates columns of statistical uncertainty as a function of energy, in this case $\dot{C}_{Ta,i}$ and $\dot{C}_{o,i}$. Finally the yellow box indicates columns of derivatives of the transmission with respect to each variable as a function of energy ($\partial T / \partial k_o, \partial T / \partial k_a$, etc). Commented out by a # just above each section of data are descriptive strings.

also does not contain the same amount of information as the separate $C_x$ and $F_x$ matrices. A compact method to report all the information necessary reconstruct a covariance matrix and all of the associated derivatives is presented herein. An example file format is given in Fig. 4.5.

The file format shown in Fig. 4.5 can be used to recreate the full covariance matrix $C_y$ for the thick sample transmission measurement. It contains the systematic uncertainties ($\Delta a, \Delta b$, etc.), statistical uncertainties ($\Delta \dot{C}_{o,i}, \Delta \dot{C}_{Ta,i}$, etc.), input covariance ($\Delta a\Delta b, \Delta b\Delta a$), and derivatives for all the variables of the transmission described in Eq. 4.1. This format is comparable to that used for the Implicit Data Covariance (IDC) matrix described by Larson in the SAMMY manual [51]. Both formats are capable of storing: input covariance
of variables (in the case of the thick sample transmission the covariance between \(a\) and \(b\), the uncertainty on systematic variables, and the partial derivatives with respect to those variables as a function of energy. The “user-supplied” IDC file format, however, only includes information for systematic variables of the observable. The format proposed here contains much more information, and can be used to recreate the full covariance matrix. It should be noted that SAMMY also allows for an explicit data covariance matrix \(C_y\) but that this feature is not recommended. Listings of the derivatives and uncertainties for the validation transmission measurement can be found in Appendix E.

### 4.2 Neutron Radiative Capture

To calculate microscopic capture cross section \(\sigma_\gamma\) the following measured quantities are needed: capture rate of the sample \(r_\gamma\), background rate associated with the specific detection geometry \(r_b\), and the relative rate of neutrons striking the sample \(R_n\). The general equation relating these quantities to capture yield \(Y\) is shown in Eq. 4.18.

\[
Y = \frac{r_\gamma - r_b}{kR_n} 
\]

The normalization constant \(k\) corrects for differences in relative neutron rate and detection efficiencies. The capture yield \(Y\) is then related to the capture cross section \(\sigma_\gamma\) as shown in Eq. 4.19, where \(n\) is the sample thickness in atoms/barn and \(\sigma_t\) is the total microscopic cross section.

\[
Y = \frac{\sigma_\gamma}{\sigma_t} \left(1 - e^{-n\sigma_t}\right) 
\]

In the RPI measurement of \(^{181}\)Ta the following quantities were measured as a function of TOF: counts with the \(^{181}\)Ta sample in the neutron beam \(C_{Ta}(t_i)\), counts with no sample in the beam (open beam) \(C_o(t_i)\), counts with the reference B\(_4\)C sample in the beam \(C_{B_4C}(t_i)\), and counts with the Pb proxy sample in the beam \(C_{Pb}(t_i)\). For the sake of brevity the remainder of this section will omit the \((t_i)\) from counts symbols. The data of interest are the counts occurring by neutron capture in \(^{181}\)Ta, found in \(C_{Ta}\) along with the background counts.
4.2.1 Background

The background includes a combination of naturally occurring radiation independent of the TOF and beam associated TOF dependent contributions from off-energy neutrons and gammas. To separate counts due to neutron captures in $^{181}$Ta and in beam background contributions due to gamma scattering the “proxy sample” method was used, which is described in detail in Ref. [50]. The idea of the proxy sample method is to choose a sample that shares electromagnetic qualities with the sample of interest, but that consists of nuclei which capture neutrons as little as possible. In this way it is possible to isolate the gamma scattering from the sample of interest from the gammas occurring by neutron capture. In general, the contribution of photon scattering to the measured count rate $\dot{C}_{Ta}$ is nearly negligible at $TOF > 10 \mu s$. The proxy sample used in this experiment is Pb as it displays similar electromagnetic qualities as the Ta sample and has a cross section which consists almost entirely of neutron scattering. Due to differences in the Pb and Ta samples such as different dimensions and different photon cross-sections, the background measured by the Pb sample is scaled by a factor $k_p$ as described in Ref. [50] before being compared to the Ta. To determine the net count rate $\dot{C}_{Ta, net}$ as demonstrated in Eq. 4.20 first the open is subtracted off, and then the additional background as measured by the proxy sample. The quantities of $\dot{M} = \frac{M}{trig}$ are the monitor rates which we define as the neutrons detected $M$ per Linac trigger $trig$ by fission chambers which are operated to monitor the average beam power. The ratio of monitor rates between different samples was typically close to unity. Superscripts found in Eq. 4.20 refer to the weighting applied to each counts vector, and the count vectors have been converted to rates as shown in Eq. 4.2.

$$\dot{C}_{Ta, net} = \dot{C}_{Ta} - \dot{C}_{Ta} \frac{M_{Ta}}{M_o} - k_p \left( \dot{C}_{Pb} \frac{M_{Ta}}{M_{Pb}} - \dot{C}_{Pb} \frac{M_{Ta}}{M_O} \right) \tag{4.20}$$

4.2.2 Weighted Neutron Capture Yield

The superscripts on variables given in Eq. 4.20 specify how the counts were weighted. The process for weighting counts measured at the 45 m capture system at RPI has been thoroughly documented in Ref [50]. The essential concept, however, is as follows. To determine when neutron capture occurs in a sample, photon cascades following the capture are detected. In order to equate the detection of a cascade to a single neutron capture event, the efficiency of the detection technique must not be dependent on the cascade path. To achieve
this, previous detectors such as the 16 segment NaI detector at RPI [68], have been designed with nearly $4\pi$ geometry to detect all of the photons emitted during a cascade and have a photon detection efficiency $\eta_\gamma$ close to 100%. Other detectors have been designed with low photon detection efficiency ($\eta_\gamma << 1$) and such that the photon detection efficiency is proportional to the energy of the photon $E_\gamma$, which is referred to as the total energy detection principle [35]. An example of this type of detector is the Moxon-Rae detector [35]. Using the total energy detection principle, the detection efficiency for a capture event is, again, insensitive to the cascade pathway. The proportionality relationship between detection efficiency $\eta_\gamma$ and photon energy $E_\gamma$ is shown in Eq. 4.21.

$$\eta_\gamma = kE_\gamma$$ (4.21)

The efficiency to detect a capture event $\eta_c$ is related to the efficiencies of all the photons in the cascade, as shown in Eq. 4.22 where $m_\gamma$ is the total number of photons in a cascade.

$$\eta_c = 1 - \Pi_{i}^{m_\gamma}(1 - \eta_{\gamma,i})$$ (4.22)

If $\eta_\gamma << 1$, $\eta_c$ is approximately equal to the sum of $\eta_{\gamma,i}$ as seen in Eq. 4.23, and independent of the cascade path. It will also be approximately proportional to the total excitation energy: the sum of the neutron binding energy $S_n$ and the energy of the incident neutron $E_n$. Note that this sum is relatively constant as $S_n >> E_n$ for the energies of interest.

$$\eta_c \approx \sum_{i=1}^{m_\gamma} \eta_{\gamma,i} \approx k \sum_{i}^{m_\gamma} E_{\gamma,i} \approx k(S_n + E_n)$$ (4.23)

In the capture yield measurements of this work, the detectors employed did not behave with efficiency perfectly linearly proportional to energy. The most probable interaction of a nuclear decay photon with C\textsubscript{6}D\textsubscript{6} atoms is Compton scattering. To account for this non-linearity, the Pulse Height Weighting Technique [74] is used to relate the energy deposited in the detector $E_d$ to $E_\gamma$. Photon scattering in C\textsubscript{6}D\textsubscript{6} deposits energy $E_d$ according to some probability distribution $R(E_\gamma, E_d)$ which is referred to as the response function. This response function is related to $\eta_\gamma$, and made to be linearly proportional to energy using a weighting function $W(E_d)$ as shown in Eq. 4.24. Following the example of Borella et. al. [75] and McDermott et. al. [33] the response functions of the C\textsubscript{6}D\textsubscript{6} detectors to
varying monoenergetic photons were simulated using the Monte Carlo code MCNP 6.1. In the MCNP model, photons were simulated starting in the sample and the energy deposited by photons reaching the detector were recorded. The starting position of the photons along the sample depth (in the direction of incident neutron travel) were randomly selected from a uniform distribution. This uniform distribution of photon starting positions is analogous to experimental conditions where the sample has a low neutron cross section and neutrons are (nearly) equally likely to capture anywhere in the sample. This is true for the majority of neutron energies for $^{181}$Ta, but not at the peaks of resonances. The distribution of captures along the sample thickness will be discussed further in §4.2.5. The response functions as calculated by MCNP using the uniform distribution were used to calculate $W(E_d)$ for each of the detectors, which were applied to the recorded counts data.

$$\eta_\gamma = kE_\gamma = \int_{E_L}^{\infty} R(E_\gamma, E_d)W(E_d)dE_d$$ (4.24)

The integral in Eq. 4.24 is taken from a lower level energy discriminator $E_L$ to $\infty$ to match the data processing where events of $E_d < E_L$ are not counted. The weighted counts for an arbitrary TOF $C^w$ are now given by real counts $C(E_d)$ associated with some $W(E_d)$ based on the energy deposition of the event in question. This is shown in Eq. 4.25.

$$C^w = \int_{E_L}^{\infty} C(E_d)W(E_d)dE_d$$ (4.25)

Once the net count rate due to neutron capture in the sample is calculated, one only needs the net rate of neutrons striking the sample $R_n$ to calculate yield. The neutron count rate is calculated as shown in Eq. 4.26.

$$R_n = \frac{\dot{C}_{B_4C}^{UW} - \dot{C}_o^{UW} M_{B_4C}}{Y_{B_4C}}$$ (4.26)

The count rates in Eq. 4.26 used to calculate $R_n$ are not weighted (superscript $UW$) because no significant gamma attenuation of the 477 keV photon following the $^{10}B(n,\alpha\gamma)^7Li$ reaction occurs in the sample, and the weighting does not vary from capture to capture. As the $^{10}B$ cross section is well known, the expected yield of the $B_4C$ sample $Y_{B_4C}$ is calculated using the program MCNP 6.1 using a neutron tally with the tally multiplier card (FM -1 1 -5) in order to obtain the total gamma production from the $B_4C$ sample. This is important
for thick samples such as the 1.3 cm thick sample used in this experiment due to the effect of secondary captures of neutrons that have first scattered in the sample. Finally neutron capture yield from the Ta sample can be calculated as in Eq. 4.27.

\[ Y_{Ta} = f_n \frac{\dot{C}_{Ta,net}}{R_n} \]  

(4.27)

The factor \( f_n \) in Eq. 4.27 is applied to normalize the weighted Ta count rate \( \dot{C}_{Ta} \) to the net neutron rate \( R_n \) striking the sample. The normalization constant \( f_n \) is necessary as the absolute count rate is not known, only the shape of the count rate \( \dot{C}_{Ta} \).

### 4.2.3 Normalization of Capture Yield

Normalization of TOF capture yield measurements of the past have often been determined by a low energy saturated resonance; where the count rate at the peak of the saturated resonance can be compared to the net neutron rate \( R_n \). As the TOF dataset in question does not have a saturated low energy resonance with which to normalize, it was necessary to explore other methods. In a procedure similar to that which has previously been employed by several authors including Barry [76] and Trbovich [77], resonances observed in the transmission measurement made at 100 meters were fit using SAMMY to determine the neutron width \( \Gamma_n \) and the capture width \( \Gamma_\gamma \) and used to normalize the capture yield. The final fit with the 100 m transmission for the 6 mm thick sample is shown in Fig. 4.6.
Figure 4.6: The experimental transmission measured at 100 m for a 6 mm thick Ta sample along with the fitted theoretical curve produced by SAMMY 8.1. The fitted parameters from these two resonances are used to normalize the capture measurement at the 45 m capture detector. The deviation of the theoretical curve from the data at the peak of the 208 eV resonance is due to the local variation of the sample background away from the smooth theoretical sample background function (Eq. 4.7). Assuming that the theoretical transmission is correct, and modifying the sample background $k_{Ta} \hat{B}$ for the experimental transmission to match the theory results in an increase on the normalization of the yield data of approximately 2.8%.

The $\Gamma_\gamma$ and $\Gamma_n$ resonance parameters that were fit to the transmission data are used to: recreate a theoretical capture yield using SAMMY, and calculate a normalization of the experimental capture yield which minimizes the $\chi^2$ in the space occupied by the resonances. The $E_\lambda$ parameters are allowed to vary to prevent the uncertainty in $E_\lambda$ from affecting the normalization. Only the normalization and $E_\lambda$ values are permitted to vary during the SAMMY fit. The capture yield based on the transmission fitted parameters (red curve) and the capture yield fit where the normalization and $E_\lambda$ values are varied (blue) are shown alongside the capture yield measurement for the 2 mm thick Ta sample in Fig. 4.7.
Figure 4.7: The experimental capture yield measured at 45 m for a 2 mm thick Ta sample along with the initial and fitted theoretical curves produced by SAMMY 8.1. The initial theoretical curve is from the parameters found by fitting the measured transmission of the 1, 3, and 6 mm Ta samples at 100 m. The fitted theoretical curve in this figure comes from allowing the normalization of the yield and the energy of the capture yield resonances to vary in the SAMMY 8.1 fit.

As the transmission measurement is mostly providing information only for $\Gamma_n$, this method depends on the capture yield being sensitive to $\Gamma_n$. The resonances used were found at $E_\lambda = 204,208$ eV, with $\Gamma_n = 2.89, 9.18$ meV and $\Gamma_\gamma = 68.3, 76.9$ meV respectively. For each of these $\Gamma_\gamma > > \Gamma_n$ and the capture area $A_\gamma$ and capture yield shape are sensitive to the neutron width. This can be illustrated using the simple Breit-Wigner equation for a single resonance and making the same assumptions as Bell and Glasstone (Ch. 8) [78] for the relation of area under the cross section resonance to the integration of cross section from $E_1$ to $E_2$. This is shown in Eqs. 4.28, and 4.29 (variables defined in §2). Using the SAMMY program to fit a normalization results in an uncertainty on the normalization $\Delta f_n \approx 4\%$.

$$A_\gamma = \int_{E_1}^{E_2} \sigma_\gamma dE = \int_{E_1}^{E_2} \left\{ \sigma_0 \frac{\Gamma}{\Gamma_\gamma} \left( \frac{E_\lambda}{E} \right)^2 \frac{1}{1 + \frac{4}{\Gamma_\gamma^2} (E - E_\lambda)^2} \right\} dE \quad (4.28)$$
\[ A_\gamma \propto g \frac{\Gamma_n \Gamma_\gamma}{\Gamma_n + \Gamma_\gamma} \]  

(4.29)

\[ A_\gamma \propto g \frac{\Gamma_n \Gamma_\gamma}{\Gamma_n + \Gamma_\gamma} \approx g \Gamma_n \quad , \quad (\Gamma_\gamma \gg \Gamma_n) \]  

(4.30)

### 4.2.4 Uncertainty and Correlation

The calculated uncertainty on the yield is taken to be the quadrature sum of the errors found in the constituents of the full yield equation, which is given explicitly in Eq. 4.31.

\[ Y_{Ta} = f_n \frac{\dot{C}_{Ta} - \dot{C}_{o} \frac{M_{Ta}}{M_{o}} - k_p \left( \frac{\dot{C}_{P} \dot{M}_{Ta}}{M_{P} M_{o}} - \frac{\dot{C}_{P} \dot{M}_{o}}{M_{P} M_{o}} \right)}{\frac{\dot{C}_{UW} - \dot{C}_{o} \frac{M_{B} C_{o}}{M_{o}}}{M_{B} C_{o}}} \]  

(4.31)

The uncertainty in the yield \( \Delta Y_{Ta} \) is given in Eq. 4.32 where \( x \) is a stand in for the different variables of Eq. 4.31. It should be noted that the fractional uncertainty for the weighted count rates is taken as equivalent to the fractional uncertainty in the un-weighted count rates plus additional uncertainty do to the weighting \( \Delta W(E_d) \). As the weights are determined from Monte Carlo calculations, the statistical uncertainty can be arbitrarily small. To estimate a realistic \( \Delta W(E_d) \) the average difference between experimental response functions and calculated response functions was calculated to be 3.78% for the C\(_6\)D\(_6\) system by McDermott [50]. The uncertainty in the weighting method is discussed further in Ref. [50].

\[ \Delta Y_{Ta} = \sqrt{\left( \frac{\partial Y_{Ta}}{\partial C_{Ta}} \Delta C_{UW} \frac{M_{Ta}}{M_{o}} \right)^2 + \left( \frac{\partial Y_{Ta}}{\partial x} \Delta x \right)^2 + ...} \]  

(4.32)

In practice the full covariance of the yield over energy \( \langle Y_i Y_j \rangle \) is not used in fitting codes such as SAMMY 8.1, though the process of uncertainty propagation as described in §4.1.3 can be applied to the yield as well. Some of the reason covariance treatment is limited in SAMMY is due to the problem of Pele's Pertinent Puzzle (PPP) (this is well documented in Ref. [79]). PPP is essentially a phenomenon that results in unexpected mean values for experimental data using statistical and systematic uncertainties. The correlated uncertainties introduced from the normalization \( f_n \), however, can be propagated to the fitted resonance parameters using SAMMY 8.1. In this case, the \( \approx 4\% \) uncertainty from the normalization discussed in §4.2.3 was given to SAMMY as an input parameter before the Bayesian fit was performed.
It should be noted that there was also correlation introduced by using the MCNP calculated $Y_{BaC}$ to determine the net neutron rate $R_n$. The $Y_{BaC}$ normalization correlated the $^{181}$Ta yield to the $^{10}$B cross section which was assumed to be known within $\approx 1\%$ in the energy range of interest [80]. Short range correlations were introduced from Savitsky-Golay smoothing of the backgrounds $C^a_{Ta}, C^P_{Pb}, C^P_{o}$.

4.2.5 Photon Attenuation

In typical capture yield experiments samples are chosen to be as thin as possible to reduce the complexity of the measured count rate in the experiment. The sample may be chosen to be thicker, however, in order to increase the capture rate and reduce the statistical uncertainty in the measurement. For the RPI measurement, a thick sample was used and required the appropriate corrections to the SAMMY 8.1 fitting process. The complexities introduced with a thick sample include neutron multiple scattering, and capture photon attenuation. The phenomenon of neutron multiple scattering is when upon the first interaction of a neutron with a nucleus the neutron scatters from the nucleus and then upon 2$^{nd}$, 3$^{rd}$, etc... interaction the neutron is captured, and a photon is detected. This process has been well modeled by the SAMMY 8.1 code. The process of photon attenuation was not included in the SAMMY 8.1 framework before this work. Part of the present Ph.D. work (Brown) involved incorporating photon attenuation corrections into the SAMMY framework based on the initial work of McDermott [50].

The process of photon attenuation (as pertaining to capture yield measurements) is when, following a neutron capture, a photon emitted from a nucleus interacts in the capture sample before escaping or is absorbed in the capture sample. The probability of a photon escaping the sample and depositing energy in the detector depends on the length of the path the photon must travel to the detector and materials it must travel through. If a neutron penetrates deeply into a sample before being captured, the photon is less likely to escape than if the neutron had been captured at the face of the sample (for back angle detectors with respect to the neutron beam). This is illustrated generically in Fig. 4.8 where photons are emitted at different average depths according to the energy of the neutron which caused them. In the RRR this presents a problem as the cross section (and therefore neutron capture depth) fluctuates violently as a function of energy. Along the profile of a neutron resonance, at the peak of a resonance neutrons are more likely to capture at the face of the
Figure 4.8: Illustration of a neutron that was collimated toward a sample, captured, and subsequently emitted a photon. Each exponential neutron flux profile across the sample thickness corresponds to the cross section found on the profile of the resonance. At the peak of a resonance in energy space, the neutron is more likely to capture at the sample surface and the emitted photon more likely to escape the sample and interact with the gamma ray detector. On the wings of a resonance a neutron is more likely to penetrate further into the sample, and the emitted photon less likely to escape.

sample, and in the wings of a resonance neutrons are more likely to penetrate deeper into the sample. This phenomenon distorts the shape of the observed resonances in the experimental data; to properly model the experimental data with SAMMY, the theoretical shape must be corrected.

4.2.5.1 Modeling the Photon Attenuation Correction

To correct the shape of the theoretical curve to fit the attenuated experimental data, a simple analytical model was chosen and fit to discrete points determined by detailed MCNP simulations. The analytical model is given in Eq. 4.33, where $k_a$ is the correction, $\mu$ is the photon attenuation coefficient, $x'$ is distance along the sample thickness, $Y_{at}$ is the photon attenuated primary yield, and $Y_0$ is the un-attenuated primary capture yield. $\Sigma_{\gamma} = N\sigma_\gamma$ is the macroscopic capture cross section.

$$k_a(\sigma_t) = \frac{Y_{at}}{Y_0} = \frac{\int_0^x \Sigma_{\gamma} e^{-\mu x'} e^{-N x' \sigma_t} dx'}{\int_0^x \Sigma_{\gamma} e^{-\mu x'} dx'} \tag{4.33}$$

Notice that the correction approaches unity for increasing values of $\sigma_t$, meaning there is hardly any correction for surface capture. It is important to keep in mind that the count
rate has already been weighted (see §4.2.2) by the efficiency of a given $E_\gamma$ to deposit energy $E_d$, where photons are simulated as coming from a uniform distribution of captures occurring over the thickness of the sample. This first weighting, which occurs event-by-event for the deposited energy $E_d$, inherently includes the efficiency for photons to escape the sample over a uniform thickness and is applied to the data in the same fashion over all energies. This is desirable as it mimics reality quite well in the URR where the cross section is smaller and contains less variation than the RRR. The first weighting, however, increases the count rate at the peak of resonances too much because it assumes the efficiency (based on uniform distribution) is less than the real efficiency (surface capture distribution). The second weighting ($k_a(\sigma_t)$) is again accounting for the efficiency of photons to escape the sample but now accounting for the cross section dependency of neutron capture distributions in the sample (exponential). The second weighting is applied to a theoretical curve to match the attenuated data. As neutron capture yield is often normalized to a saturated resonance where the yield is very close to 1, the $k_a(\sigma_t)$ correction seeks to leave the peak of the resonance unchanged and decrease the theoretical yield in the wings. This is opposed to increasing the yield in the peak of the resonance, leaving the wings unchanged, and then re-normalizing the yield to the new peak value of the saturated resonance.

For Eq. 4.33 it was assumed that only primary captures are occurring, and that the photon exits the sample precisely opposite to the vector in which the neutron entered the sample (orthogonal to the face of the sample). This assumption is justified as the photon attenuation is the quantity being fitted by a three-dimensional model. For example, one could just as easily define the distance of photon travel as $x' \cos(\theta)$ where $\theta$ is the angle between the direction of neutron travel and direction of photon travel. But this defined distance would still be fitted to the simulated model which accounts for geometry. It should be noted that Eq. 4.33 is valid for $\sigma_\gamma / \sigma_t \approx 1$, due to the assumption that the distribution of captures along the distance $x'$ is exponential. If e.g., $\sigma_t = \sigma_s + \sigma_\gamma$ and the scattering cross section $\sigma_s$ is greater than $\sigma_\gamma$, the first interaction in the sample is very likely to be scattering, making the distribution of captures along the sample depth significantly affected by the phenomenon of neutron multiple scattering. Moving forward with these assumptions the integration in Eq. 4.33 was carried out, and $\mu x$ was replaced with a fit parameter $C_1$ as shown in Eq. 4.34 (in this notation $N_x = n \text{[barn}^{-1}]$).
\[ k_a(\sigma_t) = \frac{1}{1 + C_1 \frac{n\sigma_t}{\sigma_t} \left( 1 - e^{-n\sigma_t} \right)} \]  

(4.34)

4.2.5.2 Fitting the \( C_1 \) Parameter

The function \( k_a(\sigma_t) \) was then fit to MCNP simulations for multiple values of \( \sigma_{t,i} \). For each \( \sigma_{t,i} \) there is a probability distribution \( e^{-N_x'\sigma_{t,i}} \) along the thickness \( x' \) for a neutron to capture in the sample. Each distribution corresponding to discrete values of \( \sigma_{t,i} \) was used to model the starting position of simulated photons along the sample depth. This is shown in Fig. 4.8, where it is illustrated that for larger cross section, such as at the peak of a resonance, the exponential probability decays faster, and for smaller cross section the exponential probability becomes closer to a uniform distribution along the sample depth.

When simulating the capture system, it is important to use the proper energy spectrum of photons. MCNP 6.1 was not capable of simulating the discrete values of photons in the energy spectrum of a typical capture gamma cascade, so an energy spectrum must be given to MCNP. This spectrum is obtained by a DICEBOX [81] simulation. DICEBOX is a Monte Carlo code written by Becvar which simulates gamma cascades in complex nuclei. The compound nucleus \(^{182}\text{Ta}\) de-excites with a cascade of gammas with energy probabilities shown in Fig. 4.9, where the DICEBOX calculation (blue) is compared to the Evaluated Nuclear Structure Data File (ENSDF) evaluation (green) published by the NNDC.
Figure 4.9: The energy spectrum of cascade photons occurring during the de-excitation of a compound $^{182}$Ta nucleus. This spectrum (blue) was generated using the Monte Carlo code DICEBOX [81] by Becvar which uses the RIPL and EGAF databases as input. The DICEBOX spectrum is compared with the ENSDF values (green) published by the NNDC. The red line at 0.150 MeV indicates the lower level discriminator (LLD) of the experimental analysis. The fraction of events above the LLD is $\approx 76\%$.

In the MCNP simulations, the DICEBOX energy spectrum is used to determine the energy probability distribution of the simulated photons. The discrete values of $\sigma_{t,i}$ are used to determine the distribution of simulated photon depth in the sample, and the quantity tallied is the energy deposited in detectors surrounding the sample. For each increasing value of $\sigma_{t,i}$, the exponential distribution of neutron transmission (and therefore captures) becomes more heavily weighted towards the face of the sample. The simulated correction $k_{a,sim}(\sigma_{t,i})$ is calculated from the total energy deposited in the detectors for each discrete value $n\sigma_{t,i}$ ([unitless]) simulated in MCNP which are normalized to the total energy deposited for the strongest value of $n\sigma_{t,i}$ as seen in Eq. 4.35.

$$k_{a,sim}(\sigma_{t,i}) = \frac{\sum E_d}{\text{max}(\sum E_d)}$$ (4.35)
This results in all values of simulated $k_{a,sim}(\sigma_{t,i})$ being less than or equal to 1.0. A least squares fit was performed to fit the values $k_{a,sim}(\sigma_{t,i})$ to Eq. 4.34 allowing the value $C_1$ to vary. The discrete simulated values of $k_{a,sim}(\sigma_{t,i})$ and the function from Eq. 4.34 fitted to those values for sample thicknesses of 1 and 2 mm is shown in Fig. 4.10.

![Figure 4.10: The discrete values $k_{a,sim}(\sigma_{t,i})$ for correction factor $k_a$ as determined by Monte Carlo calculation using MCNP are plotted with Eq. 4.34 fitted to those values.](image)

This photon attenuation effect is important for strong resonances measured in thick samples, as it is capable of changing the fitted resonance parameters needed to match the experimental data. A good example of this is the 6 mm Ta neutron capture yield dataset ($C_1 = 0.5545 \pm 0.114$) for the 4.28, 10, and 14 eV resonances in $^{181}$Ta as measured by McDermott et. al [50]. The high density and high proton number of Ta make it very likely for a photon to interact with a sample. The 4.28, 10, and 14 eV resonances in $^{181}$Ta are shown in Fig. 4.11, along with the SAMMY 8.1 theoretical prediction of the yield based on JEFF-3.3 parameters and the same theoretical curve multiplied by the correction $k_a$. 
Figure 4.11: The experimental data measured by McDermott [50] for the first 3 resonances in $^{181}$Ta. Plotted in solid lines are the theoretical curves as calculated by SAMMY 8.1 using JEFF-3.3 resonance parameters as input (blue line), and the same theoretical curve multiplied by the correction $k_a$ (red line).

The $k_a$ correction has now been implemented into the SAMMY program as part of the present Ph.D. thesis (Brown) and is expected to be incorporated in the next version release. SAMMY generates three separate yields, $Y_0$, $Y_1$, and $Y_2$. These yields respectively correspond to: the primary yield, yield following a single scattering event in the sample, and yield following more than one scattering events in the sample. The correction was applied to the total yield as $k_a(Y_0 + Y_1 + Y_2)$. It should be kept in mind, however, that the capture distributions for $Y_1$ and $Y_2$ are more uniform across the sample thickness than $Y_0$, making the correction less realistic. The analytical derivative of the modified yield with respect to the resonance parameters must be calculated for SAMMY to perform the Bayesian fitting process. Using the chain rule the derivative is shown in Eq. 4.36 where $u$ is the reduced resonance parameter. The derivative with respect to $\sigma_t$ is shown in Eqs. 4.37, 4.38, 4.39, and 4.40.

$$\frac{\partial (k_a Y)}{\partial u} = \frac{\partial k_a}{\partial \sigma_t} \frac{\partial \sigma_t}{\partial u} Y + k_a \frac{\partial Y}{\partial u}$$  \hspace{1cm} (4.36)
\[ \partial k_a / \partial \sigma_t = \frac{\alpha - \beta}{\tau} \]  

(4.37)

\[ \alpha = \frac{(1 - e^{-N\sigma_t}) \left( e^{-(N\sigma_t + C_1)} N (1 + \frac{C_1}{N\sigma_t}) + C_1 (1 - e^{-(N\sigma_t + C_1)}) \right)}{(1 + \frac{C_1}{N\sigma_t})^2} \]  

(4.38)

\[ \beta = \frac{Ne^{-N\sigma_t} (1 - e^{-(N\sigma_t + C_1)})}{1 + \frac{C_1}{N\sigma_t}} \]  

(4.39)

\[ \tau = (1 - e^{-N\sigma_t})^2 \]  

(4.40)

The derivatives of \( Y \) and \( \sigma_t \) with respect to the reduced resonance parameters in Eq. (4.36) have been implemented in SAMMY in the past, and can simply be utilized. They are described in the SAMMY manual [51].

Fitting the data with the properly corrected theoretical yield can be seen in Fig. 4.12. While the fitted curves do not deviate significantly from each other, the fitted value of \( \Gamma_n \) for the 4.28 eV resonance (for reference, JEFF-3.3: \( \Gamma_n, \Gamma_\gamma = 3.2, 53.0 \text{ meV} \)) changes by 28% based on whether the SAMMY program applies the correction \( k_a \).

Figure 4.12: The experimental data measured by McDermott [50] for the first 3 resonances in \(^{181}\text{Ta}\). Plotted in solid lines are the fitted theoretical curves as calculated by SAMMY 8.1 without the correction factor \( k_a \) (blue line), and the fitted theoretical curve as calculated by SAMMY with the correction \( k_a \) (red line).
CHAPTER 5
RESULTS & EVALUATION

In order to extract microscopic cross section from experimental neutron capture yield and transmission, measurements are compared to theoretical capture yield and transmission. In order to do this, the theoretical functions for capture yield and transmission must be modified by experimental parameters such as Doppler broadening and experimental resolution. The comparison of measured data and theory is typically done using standard codes, such as SAMMY in the RRR, and FITACS in the URR [51]. SAMMY is a Bayesian fitting code which uses $\mathcal{R}$-matrix theory [18] to model cross section and the corresponding transmission or capture yield (the application of Bayes equations are explained in detail by Larson in the SAMMY manual [51] and in general by Boas [82]). FITACS is a code written by Fritz Fröhner which was modified slightly to be incorporated into the SAMMY code. SAMMY in normal operation compares the transmission or capture yield, based on prior resonance parameters and $\mathcal{R}$-matrix theory, to corresponding user input data and then calculates the most probable resonance parameters given the user data and priors. When operating FITACS, SAMMY has similar Bayesian behavior but recreates cross section using prior average resonance parameters and Hauser-Feshbach theory [19][83].

The SAMMY and FITACS codes are well documented evaluation tools. The output of these codes are resonance parameters which can be stored in evaluated libraries. The goal of evaluation in this work, and in general, is to quantitatively describe measurements with parameters that can be used to reconstruct cross section more accurately. This is done in the resolved and unresolved resonance regions, and the new evaluation is validated by the thick-sample transmission measurement.

5.1 RRR

5.1.1 SAMMY Program

In the SAMMY code, the theoretical RRR transmission is modified by experimental parameters inherent in TOF measurements. The cross section is modified by Doppler broadening and the transmission is modified by resolution broadening. The resolution function $R(E, E')$ is a function used to mathematically represent broadening by uncertainty in the
flight path, TOF, and characteristics of the neutron producing target and detectors. The
mathematical formalism for broadened theoretical transmission $T_{th}(E)$ is shown in Eq. 5.1,
where $\sigma_t$ is the Doppler broadened cross section (the same formalism applies to the neutron
capture yield as well). As all experiments were performed at approximately 293 K, in the
proceeding equations it should be assumed that the cross section is Doppler broadened.

\[
T_{th}(E) = \int R(E, E')T(E)\,dE' = \int R(E, E')e^{-n\sigma_t}\,dE' \tag{5.1}
\]

In addition to Doppler and resolution broadening, capture yield requires multiple scat-
tering and photon attenuation corrections. Multiple scattering capture yield $Y_m$ occurs due
to a phenomenon where a neutron first scatters in the sample and then is subsequently
captured, leading to spurious capture events being recorded by the detectors. Multiple scat-
ering is particularly prevalent when employing thick capture samples in measurements. The
total expected yield $Y$ is depicted in Eq. 5.2 as the sum of the primary yield $Y_0$ and multiple
scattering yield $Y_m$. Multiple scattering is accounted for in the current release of SAMMY
(8.1).

\[
Y = Y_0 + Y_m = \frac{\sigma_u}{\sigma_t} \left(1 - e^{-n\sigma_t}\right) + Y_m \tag{5.2}
\]

Photon attenuation refers to the process where neutron capture photons interact with the
bulk sample material in which they were emitted. In a thin or low-Z sample this effect may
be negligible, but for thick high-Z samples such as $^{181}$Ta this effect can change the detection
efficiency as a function of neutron energy. This variation of detection efficiency as a function
of neutron energy can therefore change the shape of observed resonances. The correction for
photon attenuation is discussed in §4.2.5, and was incorporated into the SAMMY program
as part of the Ph.D. work presented herein.

5.1.2 RRR Results

The results of the neutron capture yield and transmission measurements have been
fitted using SAMMY in the RRR for multiple sample thicknesses using the Reich-Moore
approximation to the R-matrix theory [53]. The fit was carried out from the thermal region
to 2500 eV using measurements described in this work and previous data. The experimental
details and data reduction for the measurements done in this work have been described in
Table 5.1: The measurements used in this work to fit new resonance parameters using the evaluation tool SAMMY 8.1.

<table>
<thead>
<tr>
<th>Author</th>
<th>Sample</th>
<th>Data Type</th>
<th>Fitted Energy Region [eV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>J. Harvey</td>
<td>&quot;Thin&quot;</td>
<td>0.000167 at/b</td>
<td>Transmission</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.7-13</td>
</tr>
<tr>
<td>J. Harvey</td>
<td>&quot;Thick&quot;</td>
<td>0.002881 at/b</td>
<td>Transmission</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>4.4-2500</td>
</tr>
<tr>
<td>B. McDermott</td>
<td>2 mm</td>
<td>Capture Yield</td>
<td>1.7-300</td>
</tr>
<tr>
<td>B. McDermott</td>
<td>6 mm</td>
<td>Capture Yield</td>
<td>1.7-300</td>
</tr>
<tr>
<td>J. Brown</td>
<td>1 mm</td>
<td>Capture Yield</td>
<td>300-2500</td>
</tr>
<tr>
<td>J. Brown</td>
<td>2 mm</td>
<td>Capture Yield</td>
<td>300-2500</td>
</tr>
<tr>
<td>J. Brown</td>
<td>1 mm</td>
<td>Transmission</td>
<td>155-2500</td>
</tr>
<tr>
<td>J. Brown</td>
<td>3 mm</td>
<td>Transmission</td>
<td>155-2500</td>
</tr>
<tr>
<td>J. Brown</td>
<td>6 mm</td>
<td>Transmission</td>
<td>155-2500</td>
</tr>
</tbody>
</table>

Chapters 3 and 4, respectively. The measured data which were fit and the energy regions in which they were fit are listed in Table 5.1.

The prior information given to SAMMY were the JEFF-3.3 resonance parameters, which include 2 negative energy resonances below the fitted RRR. Six resonances above 2500 eV (unvaried from JEFF-3.3) plus a constant addition to the R-matrix $R_i^\infty$ term were used to account for “distant level” contributions as described by Fröhner [54] and in §2.1. As no reliable thermal data were available to be fitted, it was decided to constrain the thermal cross section value to that of JEFF-3.3 as it matches closely to Mughabghab’s values in the Neutron Resonance Atlas [55]. This was accomplished by using JEFF-3.3 point-wise cross section from 0.01-0.1 eV, applying arbitrarily low errors, and having SAMMY fit these data sequentially along with the real data sets. Constraining the thermal data to a reasonable value allowed for more reliable resonance parameters for the lowest energy resonances fitted in this work, though it should be noted that this method introduces an evaluation bias towards JEFF-3.3. The results of the RRR fit are shown in Figs. 5.1-5.6.
Figure 5.1: Low energy experimental data. The transmission measurements come from the Harvey datasets [34], and the capture yield measurements were made at RPI [33]. The “thin” and “thick” Harvey samples were 0.000167 and 0.002881 at/b, respectively. The lowest energy datum for the “thick” Harvey measurement was \(\approx 4.4\) eV in the middle of a resonance, and was fit down to that energy. The legend labels end in “C” or “T” to indicate capture yield and transmission respectively. It should be noted that the 4 eV resonance has the greatest impact on integral quantities for \(^{181}\text{Ta}\).

Fig. 5.1 includes the McDermott data [33][50] and the Harvey transmission data sets [34]. The McDermott data includes 2 and 6 mm samples, and provided the most well resolved capture yield (or capture cross section) data currently available for the energy region \(\approx 1\) eV to 200 eV. It should be noted that the McDermott data (in particular the 6 mm data) had to be modeled in SAMMY properly using neutron multiple scattering and the photon attenuation correction described in §4.2.5. A closer view of the 4.28 eV resonance is shown in Fig. 5.2.
Figure 5.2: Low energy resonance fitted to experimental data. The transmission measurement comes from the Harvey dataset [34], and the capture yield measurements were made at RPI [33]. The “thin” Harvey sample was 0.000167 at/b in areal density. It should be noted that the 4 eV resonance has the greatest impact on integral quantities for $^{181}$Ta. The agreement of the RPI and Harvey datasets indicates that the $k_a$ correction improved the model of the data.

The thin-sample Harvey transmission measurement is just thin enough to prevent saturation of the 4.28 resonance. This is important to properly predict $\Gamma_\gamma$ and $\Gamma_n$. The McDermott capture yield data for both sample thicknesses agrees relatively well, but discrepancies exist in the width of the resonance. These discrepancies are part of the uncertainty in the theoretical yield: uncertainty in the correction, normalization, and resolution function. The 4.28 eV capture resonance was affected more than any other resonance by the photon attenuation correction, and a more accurate measurement of the 4.28 and 10 eV resonances could be obtained with a new thin sample capture yield measurement. The fact that the 4.28 eV resonance in the Harvey transmission data and the McDermott capture data agreed relatively well with the same fitted resonance parameters lends credence to the photon attenuation correction incorporated into SAMMY.
Figure 5.3: Resonances are shown past the ENDF/B-VIII.0 RRR, and are seen to be well resolved. The legend labels end in “C” or “T” to indicate capture yield and transmission respectively.

Fig. 5.3 shows nearly all of the data sets used for the SAMMY evaluation. The McDermott capture yield data were fitted up to 300 eV, after which the capture yield data for the present work (Brown) were fitted. This energy split was chosen based on the poor signal to background ratio for the Brown capture yield data below $\approx 300$ eV, as well as the fact that the resolution of the McDermott measurement was adequate to resolve resonances up to at least 300 eV. The transmission data sets included the thicker sample Harvey data as well as the 1, 3, and 6 mm data sets obtained in the present work (Brown). The thick sample Harvey data were fitted from $\approx 10$ to 2500 eV. The Brown transmission data sets were fit from $\approx 160$ to 2500 eV; the lower limit of this region was somewhat determined by the fixed Co notch that removes a significant portion of the neutron rate near the 132 eV Co resonance. JEFF-3.3 improperly labeled a resonance at 304 eV, the comparison of JEFF-3.3 and the SAMMY fit yield and transmission is shown in Fig. 5.4.
Figure 5.4: The 304 eV resonance is highlighted here by inaccurate resonance parameters published in the JEFF-3.3 evaluation. This discrepancy can be seen in the transmission and capture yield datasets. The RPI fit corrects this inaccuracy to better match the data.

The Harvey “thick” sample transmission dataset has very good statistical uncertainty, but was obtained using a sample that was quite thin (0.002881 at/b), meaning that it cannot provide much information on the effective radius and potential scattering between resonances. The thicker sample measurements by Brown compensated for this, which made the combined datasets a good estimate of the expected transmission.
Near the end of the RRR of JEFF-3.3 the experimental data are very likely to be missing levels, but the Reich-Moore R-matrix model represents the data very well. The legend labels end in “C” or “T” to indicate capture yield and transmission respectively.

Fig. 5.5 shows the highest energy RRR fitted data from the previously mentioned datasets. In this energy region it became more obvious that measurements may be missing resonances. It should be noted that, while nuclear levels may have been missed, the fitted theoretical transmission matches the measurements very well up to 2.5 keV. The end of the RRR is shown over a small range to better distinguish resonances in Fig. 5.6. The JEFF-3.3 evaluation did not include a resonance at 2460 eV, for the RPI evaluation this resonance has been added. It can be seen that the 2460 eV resonance does not match perfectly with the 2 mm thick sample capture yield data. This may indicate that greater weight is given to the transmission measurements than the capture yield measurements for that resonance. It can also be seen that the measured capture yield is lower than the theoretical capture over the energy range from 2410 to 2450 eV; this is likely due to missing resonances in addition to combined statistical uncertainty in the sample count rate and associated background measurement.
5.1.3 Resonance Statistics

Once resonance parameters were obtained through the Bayesian fitting process using the SAMMY program, the parameters were compared to the expected statistical distributions based on the current physical understanding of the nucleus. The distribution of measured average level spacing and the average reduced neutron widths were compared to the theoretical Wigner [85] and Porter-Thomas [58] distributions, which were described in §2.3 and repeated here. Wigner’s distribution is shown in Eq. 5.3 and the Porter Thomas in Eq. 5.4. Keep in mind the variable definitions, where \( \nu \) is the degrees of freedom, \( S = E_\lambda - E_{\lambda-1} \) is the level spacing, and \( \Gamma \) is the Gamma function, not a resonance width (further discussion on these distributions can be found in §2.3).

\[
p(S/D) dS = \left\{ \frac{\pi S}{2D^2} e^{-\frac{\pi s^2}{4D^2}} S \right\} dS
\]

\[
p(y) dy = \frac{\nu y^{\nu/2 - 1} e^{-\nu/2y}}{2 \Gamma\left(\frac{\nu}{2}\right)} dy, \quad 0 < y \equiv \frac{\gamma_x^2}{\langle \gamma_x^2 \rangle} < \infty
\]
The measured and theoretical Porter Thomas distributions for the widths and the Wigner distributions of level spacing are shown in Fig. 5.7, where $x_1 = S/D$ and $x_2 = \Gamma_0^0 / \langle \Gamma_0^0 \rangle$ (keep in mind that $S$ is the level spacing, $D$ is the average level spacing, and $\Gamma_0^0_n$ is the reduced neutron width for s-waves). The RPI measurement was discrepant with the theoretical distribution of the reduced neutron widths, but did not differ greatly from JEFF-3.3. The Porter Thomas distribution for the $J = 3.0$ levels had an excess of levels below $\sqrt{x_2} < 1$, and for $J = 4.0$ the distribution diverges significantly from the theoretical distribution. The measured distribution of $J = 3.0$ and $J = 4.0$ level spacings match the theory reasonably well, but there is an excess of large level widths (near $\sqrt{S/D} = 1.5$). This indicates that the measurement (and JEFF-3.3) are missing resonances, the impact of which can be tested with the validation transmission in §5.3. The energy dependence for the number of missed levels can be seen more clearly with a cumulative level plot.
Figure 5.7: For the both the level spacing and level width plots the plotted distributions are the number of resonances greater than $x_1 = S/D$ and $x_2 = \Gamma_0^1/\langle \Gamma_0^1 \rangle$, respectively. Note that the abscissa of the plots are $\sqrt{x_1}$ and $\sqrt{x_2}$. The top left and right plots show the normalized Wigner distributions for the $J = 3.0$ and $J = 4.0$ level spacing, respectively. The variable $x_1 = S/D$. The Wigner distribution derived from the RPI measurement for $J = 3.0$ had 1 level greater than $\sqrt{x_1} = 2.5$. The bottom left and right plots show the normalized Porter Thomas distributions for the $J = 3.0$ and $J = 4.0$ reduced neutron widths, respectively. The Porter Thomas distribution calculated from the RPI measurement for $J = 4.0$ contains 3 levels greater than $\sqrt{x_2} = 3.5$.

Missing nuclear levels and improper spin assignments may cause the cumulative number of levels to behave non-linearly over the energy range from 0-2.5 keV. The measured cumulative levels are compared to the inverse average level spacing in Fig. 5.8. In the resolved resonance region, the level spacing (or level density) should not vary greatly for heavy isotopes such as $^{181}$Ta. Using the cumulative levels as a guide, the region in which the level spacing behaves linearly can be observed. Obviously the observed levels from 0-2.5 keV do not behave linearly. The level spacing from 0-300 eV, however, displayed linear behavior and can be used to calculate the average level spacing for $^{181}$Ta.
Figure 5.8: The cumulative levels as a function of energy compared to the expected cumulative levels as a function of energy. The slope of the expected cumulative levels is given by $1/D$. $D$ was calculated from the low energy region of 0-300 eV as its behavior diverged from linearity shortly after 300 eV. This non-linearity could be due to missing levels.

Aggregate resonance parameter statistics show two things: resonances are being missed, and that the majority of these missing resonances have small widths. It’s possible that spin assignments of $J = 3.0$ should be changed to $J = 4.0$ to follow the expected theory better. Nuclear data are used to understand nuclear physics and to predict how nuclear applications will behave. In terms of nuclear physics, the extension of the RRR beyond 330 eV may be unwarranted. In terms of nuclear applications, however, the most important indicator of the quality of a given set of resonance parameters is how well that set performs to reproduce measured quantities. The validation transmission measurement shown in §5.3 provides feedback on the performance of the RRR parameters which have been extended to 2.5 keV, even though resonances are missing.

5.1.4 RRR Data Format

An important part of data evaluation is the method used to store the evaluated parameters so that the nuclear data community may have access to them. This was done by formatting the resonance parameters to the ENDF format [28] agreed upon by the Cross
Section Evaluation Working Group (CSEWG). This format was used by the major nuclear
data libraries that have been discussed throughout this work. A convenient feature of the
SAMMY program is that it was designed to output the final fitted parameters from an eval-
uation to the ENDF format File 2. This feature was used for this work to produce a File 2
with new fitted parameters.

It has been suggested by Larson [51] and others that the nuclear data community should
move away from the Single/Multi Level Breit Wigner (SLBW) (MLBW) formalisms to the
more rigorous Reich-Moore formalism. Despite this, there are many isotopes that remain
listed in libraries with the MLBW formalism, including $^{181}$Ta. The Reich-Moore formalism
is important for properly modeling resonance interference effects. To address this the ENDF
format includes formats that can store all the information necessary for the Reich-Moore
formalism, including the “limited R-matrix” LRF=7 format for File 2 (see Ref. [28]). The
fitted parameters in this work were fitted using the rigorous Reich-Moore formalism, and
therefore stored in a format (LRF=7) that processing codes such as NJOY21 could use to
recreate cross section properly. This is an important advancement of the $^{181}$Ta isotope, as it
brings the community one step closer to modeling the true cross section.

5.2 URR

Average parameters for the URR were derived from the fitted RRR parameters. These
include the distant level parameter $R_0^\infty$, the neutron strength function $S_i$, the average ra-
diation width $\langle \Gamma_\gamma \rangle$ and the average s-wave level spacing $D$. $R_0^\infty$ was calculated by fitting
experimental data in SAMMY for this work. The “effective” radius $R'$ can also be related
to the $R_0^\infty$ term for s-waves by Eq. 5.5 where the channel radius is given by $a_c$.

$$R' = a_c(1 - R_0^\infty) \tag{5.5}$$

A simple average over radiation widths $\left(\frac{1}{\Lambda} \sum^\Lambda \Gamma_{\gamma,i}\right)$ for all resonances $\Lambda$ was taken to
calculate an average radiation width $\langle \Gamma_\gamma \rangle$, and the average level spacing $D$ was taken as the
mean distance between resonances from 4-300 eV ($\Lambda/\Delta E$, only s-waves were observed). The
energy region $\Delta E$ over which to calculate the mean level spacing was justified by the region
over which the level spacing maintained linearity with energy. Above 300 eV, the observed
level density slowly and steadily changes. Another reason to use a low energy range for $D$ is
based on the way that FITACS calculates level density as a function of energy. Quoted from the SAMMY manual [51] the input quantity $D$ should be “the mean level spacing of the $l = 0$ resonances at $E = 0$”, which means the program expects the level spacing found very close to the neutron binding energy. The formula for the strength function as seen in the Atlas by Mughabghab has been defined in §2.1 and is repeated in Eq. 5.6 for convenience. As defined here $l$ is the quantum orbital angular momentum and $g_\lambda$ is the spin statistical factor for each resonance $\lambda$ (full variable definition in §2.1).

$$\tilde{S}_l = \frac{1}{(2l + 1)\Delta E} \sum_{\lambda} g_\lambda \Gamma_{\lambda,n}^l$$  \hspace{1cm} (5.6)

These quantities are then used as input prior information to the SAMMY implementation of the FITACS code; an example FITACS (URR SAMMY PAR file) is given in Appendix A. Average total and capture cross section data from 2-120 keV were then fit to find the most probable average resonance parameters using Bayes equations. As FITACS was designed only to fit cross sections and not neutron transmission or capture yield, the observables of $T_{exp}$ and $Y_{exp}$ must be converted to the proper cross section. This presented a problem as multiple scattering and resonance self shielding must be accounted for in the conversion to cross section. This was solved by using the Monte Carlo code SESH by Fröhner [63], which was used to calculate a self shielding correction factor for transmission data $C_T$ and a self-shielding and multiple scattering correction factor for the capture yield $C_C$. An example of the effect the self-shielding correction has on the observed total cross section for the 6 mm Ta sample is shown in Fig. 5.9.
Figure 5.9: The measured average total cross section is shown corrected (black) and uncorrected (blue) for resonance self-shielding by SESH. The JEFF-3.3 evaluated cross section is shown as a reference for the expected cross section prior to this measurement.

The self-shielding correction factors from SESH are used to correct the experimental transmission and capture yield as shown in Eqs. 5.7 and 5.8, where $\sigma_{\gamma,\text{sim}}$ is the “average effective capture cross section” as calculated by SESH. The average total cross section $\langle \sigma_t \rangle$ comes from the Hauser Feshbach theory, and the average transmission $\langle e^{-n\sigma_t} \rangle$ is obtained by a Monte Carlo simulation for a given transmission sample. $Y_{\text{sim}}$ is the simulated yield as calculated by SESH. Recall that $n$ is the areal number density in at/b.

$$
\langle \sigma_{t,\text{exp}} \rangle = \frac{-1}{n} \ln \left( \frac{\langle T_{\text{exp}} \rangle}{C_T} \right), \quad C_T = \frac{\langle e^{-n\sigma_t} \rangle}{e^{-n\langle \sigma_t \rangle}}
$$

$$
\langle \sigma_{\gamma,\text{exp}} \rangle = \frac{\langle Y_{\text{exp}} \rangle}{nC_C}, \quad C_C = \frac{\langle Y_{\text{sim}} \rangle}{n\langle \sigma_{\gamma,\text{sim}} \rangle}
$$

The capture cross section formula shown in Eq. 5.8 is the “thin sample” approximation, and is the first term in a Taylor series expansion of the exponential term in Eq. 5.9 divided by the correction factor.

$$
\sigma_{\gamma} = Y\sigma_t (1 - e^{-n\sigma_t})^{-1}
$$
SESH is not, however, a fitting code. Before a correction can be calculated the average parameters must already be known. Average parameters such as the average radiation width \( \langle \Gamma_{\gamma,l} \rangle \), average level spacing \( D_l \), neutron strength function \( \tilde{S}_l \), inelastic neutron strength function \( S'_l \) (set to 0), and the effective scattering radius for each partial wave \( l = 0, 1, 2... \) must be given as input. An example of a SESH input file used for this work is shown in Appendix A. SESH uses these average parameters and input sample geometry to calculate effective transmission and capture yield in a Monte Carlo calculation. As it is not a fitting code, the process of using SESH was iterative, where the average parameters used for SESH and SAMMY/FITACS must converge. The iterative process is illustrated in Fig. 5.10, where iteration continued until the SESH calculated correction factor changed by < 1% between iterations.

![Diagram of iterative process](image)

**Figure 5.10:** A diagram of the iterative process for fitting grouped total and capture cross section in the URR using the SESH program and FITACS program in SAMMY. The data are corrected by evaluated parameters to start, and then the fitted parameters correct the data to be re-fitted until a convergence has been found for the correction factor calculated by SESH. Usually 2-3 iterations achieve convergence.

To begin, RRR parameters were used to calculate URR average parameters and input to SESH to calculate a correction. Then the corrected cross sections are input to FITACS to obtain fitted URR average parameters. These parameters are fed back to SESH for the next iteration, typically for not more than 2 iterations. The fitted average cross sections in the URR following this iteration are shown in Figs. 5.11 and 5.12.
Figure 5.11: The measured average capture cross section is corrected for multiple scattering and self-shielding by SESH and fitted using SAMMY/FITACS. The uncertainty on the measured data comes from statistical uncertainty in the count rate and systematic uncertainty in the flux normalization and background subtraction. It was assumed that the flux, background and measured capture rates were not correlated. The dashed curve represents a theoretical cross section using the average parameters calculated from the RRR measurement.

The uncertainty on the total and capture cross sections include statistical and systematic uncertainty, but assume that the uncertainty on the input variables is uncorrelated (e.g., uncertainty on \( n \) is assumed to be uncorrelated to uncertainty on \( \langle T_{exp} \rangle \) in Eq. 5.7). Along with the final curves for total and capture cross sections, the dashed curves for the RRR fitted parameters are shown. The fact that the URR theoretical cross sections using average parameters directly calculated from the RRR measurement (dashed red curves) passed within the error bars of the URR measured data provided further validation of the choices made in the evaluation of the RRR, including the average level spacing \( D = 4.17 \) eV. It should be kept in mind that the uncertainty on the total cross section as derived from transmission is \( \propto \frac{1}{n} \), meaning the thinner samples (e.g., the 1 or 3 mm \(^{181}\)Ta samples) include greater uncertainty for the total cross section.
Figure 5.12: The measured average total cross section is corrected for self-shielding by SESH and fitted using SAMMY/FITACS. The dashed curve represents a theoretical cross section using the average parameters calculated from the RRR measurement. It should be noted that the uncertainty in the measured total cross section includes statistical uncertainty from the count rate and systematic uncertainty deriving from the background subtraction method described in §4.1.2.

A summary of the average parameters from RPI measurements is compared to those of Mughabghab [55] and McDermott [33] in Table 5.2. The average parameters in the column labeled as RRR are calculated from the fitted RRR measurements described previously. The RRR parameters shown in Table 5.2 determine the prior probabilities for Bayes equations used in the FITACS code, and the URR parameters are the result of the posterior most probable parameters due to the priors and average experimental data. The URR initial (I) parameters come from the fitted RRR parameters where available, however, $\tilde{S}_l$ for $l = 1, 2$ is taken from Mughabghab, the $\langle \Gamma_{\gamma, l} \rangle$ for $l = 0$ is repeated for $l = 1, 2$, and the $R_l^\infty$ for $l = 1, 2$ is set to zero with a large uncertainty. The URR final (F) parameters are taken directly from the fitted parameters output from the FITACS code.

The difference between the accepted value of $a_c$ by RPI (Brown) and McDermott should be noted. This difference can be somewhat compensated by the $R_l^\infty$ terms, but still produces subtle differences in the final cross section. The $a_c$ value is used to calculate
Table 5.2: Various average level parameters used to describe the URR from 2-120 keV. The RPI parameters taken from the resonance parameters fit with SAMMY are listed for those which could be measured. The initial parameters used for the URR fit in FITACS (marked by “I”) take the RRR average parameters where possible; see the text for discussion of the other parameters. The final fitted parameters in the URR (marked by “F”) are the final output from the FITACS fit. The average parameters from McDermott [33] are listed, as well as the average parameters found in the Atlas of Neutron Resonances [55] by Mughabghab. McDermott used the average level density of Mughabghab, as indicated by the *.

Subscripts in the $\langle Par \rangle$ column indicate quantum angular momentum for $l = 0, 1, 2$. Uncertainty is omitted for quantities related to $a_c$, as $a_c$ does not have a defined uncertainty.

<table>
<thead>
<tr>
<th>$\langle Par \rangle$</th>
<th>RPI (RRR)</th>
<th>RPI (URR I)</th>
<th>RPI (URR F)</th>
<th>McDermott</th>
<th>Atlas</th>
</tr>
</thead>
<tbody>
<tr>
<td>$S_0 \cdot 10^4$</td>
<td>1.84±0.11</td>
<td>1.84±0.11</td>
<td>1.57±0.03</td>
<td>1.71±0.12</td>
<td>1.74±0.12</td>
</tr>
<tr>
<td>$\tilde{S}_1 \cdot 10^4$</td>
<td>n/a</td>
<td>n/a (Atlas)</td>
<td>0.91±0.11</td>
<td>0.52±0.10</td>
<td>0.5±0.2</td>
</tr>
<tr>
<td>$\tilde{S}_2 \cdot 10^4$</td>
<td>n/a</td>
<td>n/a (Atlas)</td>
<td>1.89±0.22</td>
<td>2.6±0.6</td>
<td>2.3±0.3</td>
</tr>
<tr>
<td>$\langle \Gamma_{\gamma,0} \rangle$ [meV]</td>
<td>67.0±11</td>
<td>67.0±11</td>
<td>62.6±0.1</td>
<td>62.6±5.1</td>
<td>60.5±2.0</td>
</tr>
<tr>
<td>$\langle \Gamma_{\gamma,1} \rangle$ [meV]</td>
<td>n/a</td>
<td>67.0±11</td>
<td>42.0±0.5</td>
<td>55.0±13.1</td>
<td>n/a</td>
</tr>
<tr>
<td>$\langle \Gamma_{\gamma,2} \rangle$ [meV]</td>
<td>n/a</td>
<td>67.0±11</td>
<td>62.6±0.1</td>
<td>62.6±5.1</td>
<td>n/a</td>
</tr>
<tr>
<td>$D$ [eV]</td>
<td>4.16±0.01</td>
<td>4.16±0.01</td>
<td>4.16±0.01 (nv)</td>
<td>4.17±0.04*</td>
<td>4.17±0.04</td>
</tr>
<tr>
<td>$R_0^\infty \cdot 10^3$</td>
<td>-6.6±0.7</td>
<td>-6.6±1.0</td>
<td>-6.6±0.9</td>
<td>41±20</td>
<td>n/a</td>
</tr>
<tr>
<td>$R_1^\infty \cdot 10^3$</td>
<td>n/a</td>
<td>0.0±10.0</td>
<td>-1.3±9.9</td>
<td>13±19</td>
<td>n/a</td>
</tr>
<tr>
<td>$R_2^\infty \cdot 10^3$</td>
<td>n/a</td>
<td>0.0±10.0</td>
<td>-0.2±9.9</td>
<td>18±11</td>
<td>n/a</td>
</tr>
<tr>
<td>$R'$ [fm]</td>
<td>7.85</td>
<td>7.85</td>
<td>7.85</td>
<td>7.67±0.08</td>
<td>7.6±0.2</td>
</tr>
<tr>
<td>$a_c$ [fm]</td>
<td>7.8</td>
<td>7.8</td>
<td>7.8</td>
<td>7.92</td>
<td>n/a</td>
</tr>
</tbody>
</table>

the penetrabilities $P_l$, and is intimately bound to the final resonance parameters. In some ways this makes the two sets of parameters difficult to compare. It should also be noted that there are many suggestions for how to estimate $a_c$. In the SAMMY formalism [51] the default formula for $a_c = 1.45(1.008665 + A)^{1/3}$, in Refs. [17][28] $a_c = 1.23A^{1/3} + 0.8$, and in Ref. [55] Mughabghab defines his “interaction radius” $R = 1.35A^{1/3}$. When performing evaluations, care must be taken to define the $a_c$ which is used to calculate penetrability and shift factors as well as the effective radius $R'$ (if they are different).
5.3 Validation Transmission

5.3.1 MCNP

The validation transmission measurement was simulated using MCNP 6.1 to assess how well the transmission could be modeled by a continuous energy Monte Carlo code. The MCNP 6.1 input file for the validation transmission model is given in Appendix A. In the MCNP model neutrons are simulated all traveling in a well collimated beam 3 cm in diameter and traveling a 35.18 m flight path $L$ to the detector. For the “sample in” simulation the neutrons must pass through a 12 mm sample of Ta ($\rho = 16.92$ g/cm$^3$, thermodynamic temperature = 293.6K) at 20 m. For the “open” simulation, the sample cell was voided, which allows neutrons to pass forward to the detector. The neutrons were tallied upon crossing the detector surface, and the TOF of the neutrons were binned in time over the TOF range of $3.5 \rightarrow 500 \mu s$. To mimic the experimental resolution of the measured data, the simulation also had a 10 ns burst width for neutron emission. The experimental resolution would have to be modeled in greater detail for a high-resolution RRR transmission comparison but is negligible compared to the energy grouping of the measured validation data and corresponding simulated transmission. To compute the transmission, the ratio of the “sample in” and “open” tallies was taken as a function of TOF. The MCNP transmission was grouped in an identical fashion to the measured data to provide a fair comparison.

5.3.2 Results

In a TOF neutron transmission measurement the count rates $\dot{C}_{Ta}(t_i)$ and $\dot{C}_o(t_i)$ are always measured in a time bin $t_i$ of some finite width $\Delta t$. To mimic URR measurement conditions (poor energy resolution), we can simply group the counts into larger bins $t_i$. This is done in such a way that each bin contains multiple resonances. In this way we increase the total fluctuation of the cross section in the measured bin and pronounce the self-shielding effect. For the thick-sample transmission measurement, it was ensured that a statistically significant number of resonances were contained in each group. To mimic a TOF measurement with large time bins $t_i$, the counts as a function of time $C_{Ta}(t_{i,raw})$ and $C_o(t_{i,raw})$ were grouped in time bins of minimum width $\Delta t$ as calculated in Eq. 5.10.
\[ \Delta t = t_i - t_{i-1} = \left( \frac{L}{c} \sqrt{1 - \frac{1}{\left(\frac{E_i}{m_n c^2} + 1\right)^2}} \right) - \left( \frac{L}{c} \sqrt{1 - \frac{1}{\left(\frac{E_{i-1}}{m_n c^2} + 1\right)^2}} \right) \]  

(5.10)

In Eq. 5.10, \( \Delta t \) is the difference in time between energy \( E_{i-1} \) and energy \( E_i \) which is defined as \( \Delta E \) (the conversion from energy \( E_i \) to TOF here is using the relativistic relation). This energy bin \( \Delta E \) is selected by ensuring a statistically significant number of resonances \( N_{\text{res}} \) reside in each bin in the grouped TOF. For the thick-sample transmission measurement, a minimum 50 resonances are grouped in each bin and the evaluated average level spacing \( D = 4.17 \text{ eV} \), resulting in a minimum \( \Delta E = N_{\text{res}} D = 208.5 \text{ eV} \). After the counts have been grouped, they were converted to count rate, background subtracted, and transmission was calculated as described in §4.1. The grouped transmission is shown in Fig. 5.13.

![Figure 5.13](Image)

**Figure 5.13:** The grouped thick-sample transmission measurement in red, along with the MCNP calculated transmission using various evaluated cross section libraries.

In the comparison of the ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 libraries to the thick-sample transmission measurement, there are obvious discrepancies. ENDF/B-VIII.0 ends the RRR at 330 eV and does not properly model the structure found in the transmission from 330→2500 eV. The ENDF/B-VIII.0 evaluation has the URR from 330→5000 eV, which
allows the average transmission to be corrected for resonance self-shielding up to 5 keV. All energies above 5 keV use the smooth average cross section listed in ENDF format File 3. The abrupt change in the transmission at 5 keV using the ENDF/B-VIII.0 library is caused by the transition from the URR model to File 3 smooth cross section, in addition to improper interpolation.

JEFF-3.3 and JENDL-4.0 have identical RRR evaluations; this is represented by the two transmission calculations being identical for the simulated transmission up to 2.5 keV. The two libraries have differing evaluations of the URR: JEFF-3.3 used average resonance parameters in File 2 to recreate the cross section, whereas JENDL-4.0 listed smooth cross section in File 3 and corrected the smooth cross section for resonance self-shielding using the average parameters in File 2. In the lower URR JENDL-4.0 was closer to the measured transmission, and in the higher energy URR JEFF-3.3 was performing better. Based on the level of agreement of the two libraries with the measured transmission in the URR, either method of modeling the URR could be employed with success. The greatest discrepancy between the measurement and the JEFF-3.3/JENDL-4.0 libraries occurs between 2.5→20 keV.

This was partly due to inaccurate level spacing parameters listed in JEFF-3.3, which results in an inaccurate resonance self-shielding correction. The evaluated level spacing given in JEFF-3.3 was greater than the true level spacing, the fluctuation from the average cross section was over-predicted, and the resulting transmission was greater than the true transmission. This effect diminished as energy increased as the widths of the resonances overcame the spacing between the resonances to effectively average the true cross section. The discrepancy could also be attributed the incorrect value of $R'$ in the URR. JEFF-3.3 and JENDL-4.0 both list 7.8, but the RPI evaluation would suggest that $R' \approx 7.85$ (fitted result for RRR and URR) results in better agreement to the measured data.

The uncertainty on the thick-sample transmission was taken to be the square root of the diagonal of the $\langle T_i T_j \rangle$ covariance matrix. The propagation of the uncertainties and covariance for the transmission has been described in detail in §4.1.3. The covariance matrix was converted to a correlation matrix and plotted in Fig. 5.14. The fractional error in the transmission measurement is plotted above the correlation matrix, and to the right is the corresponding transmission.
Figure 5.14: The correlation matrix for the $\langle T_i T_j \rangle$ covariance matrix for the thick-sample transmission measurement. Plotted above the correlation matrix is the fractional uncertainty in the transmission measurement, and to the right of the correlation matrix is the corresponding transmission. The two dashed peaks in the fractional uncertainty (up to $\approx 5\%$) are caused by Al resonances at 35 and 88 keV.

As expected, the fractional uncertainty in the transmission is greatest where large resonances in structural Al occur (35 and 88 keV resonances were identified with a dashed line in the top plot of Fig. 5.14). The Al resonances significantly reduce the measured signal at the isolated energies of approximately 6, 35, and 88 keV, resulting in uncertainties in the transmission as high as $\approx 5\%$. The degree of the energy-to-energy correlation of the transmission measurement is due to the monitor normalization and background subtraction method. The same normalization and fitted parameters are used to model the time dependent background for all bins $t_i$ which, along with a constant room background, is subtracted from the uncorrelated count rates.
5.3.3 Improving the Model

Once it was found that none of the existing evaluated libraries could reproduce the thick-sample transmission measurement over all energies using a continuous energy Monte Carlo calculation, the next task was determining a set of parameters and/or modeling method that would perform better. Several different methods were tested to ascertain whether re-evaluation of $^{181}$Ta transmission and capture yield data might improve a continuous energy Monte Carlo calculation of the validation transmission. The simplest change made was modifying the existing RRR parameters and average parameters in the URR for JEFF-3.3. The other methods investigated here were the extension of the RRR to 4 keV and modeling the URR cross section with multiple sets of resonance parameters in energy regions: $2\rightarrow10$ keV, $10\rightarrow45$ keV, and $45\rightarrow120$ keV. The process of evaluating data to fit resonance parameters, processing the parameters to continuous cross section, and performing Monte Carlo calculations is illustrated in Fig. 5.15.

![Diagram of the work flow for evaluating measured data and testing how well the new evaluation predicts neutron transmission.](image)

**Figure 5.15:** A description of the work flow for evaluating measured data and testing how well the new evaluation predicts neutron transmission. The measured data are fitted for resonance parameters, formatted into ENDF/B format files, processed by the NJOY21 program to an ACE file, and finally used by MCNP in a Monte Carlo calculation.

The evaluation data set was input to SAMMY and FITACS to generate a set of resonance parameters for the resolved and unresolved resonance regions, respectively. The
SAMMY 8.1 program includes functions to output ENDF/B format File 2. The ENDF/B formatted parameters from SAMMY 8.1 were placed into the ENDF/B format JEFF-3.3 evaluated file. This modified file was then processed by NJOY21. NJOY21 recreated the point-wise cross section based on the modified resonance parameters in the RRR and URR (along with additional cross section information throughout the file), Doppler broadened the cross section, and formatted the continuous energy cross section into “A Compact ENDF” (ACE) format file that can be read by MCNP 6.1. ACE files can then be used by MCNP to sample the cross section at each energy in the Monte Carlo calculation. This process was used to validate the various evaluations presented in this work.

5.3.3.1 Modification of Existing Parameters

The most minimal change to an evaluation that could be made was to replace existing parameters with newly fit parameters and change nothing else. As the JEFF-3.3 evaluation arguably performed the best among the tested evaluations, it was chosen to be the base evaluated file on which to make modifications. In a recent publication by McDermott et. al [33], prior to the RRR and URR evaluation shown in §5.1 and §5.2, a set of average resonance parameters was fit using SAMMY/FITACS to a Ta capture yield measurement. The McDermott measurement was an Fe filtered beam capture yield measurement. This type of measurement is a low background method to measure capture yield at the isolated energies that neutrons are transmitted through a thick slab of Fe. As a preliminary test of the method, the reported average resonance parameters by McDermott were used to replace the URR evaluation in JEFF-3.3. The modified file was used as input to NJOY21, and the NJOY21 generated ACE file was used as input to MCNP 6.1 to calculate an expected transmission. The thick-sample transmission measurement, evaluated libraries, and JEFF-3.3 modified evaluation with URR parameters reported by McDermott are shown in Fig. 5.16.
As new measurements have been made, the cross section can be re-evaluated. The evaluation of the RRR described in §5.1 provided newly fit resonance parameters from 4→2500 eV. These parameters were used to replace the existing RRR evaluation in JEFF-3.3. The evaluation of the URR described in §5.2 supplies average resonance parameters fitted from 2→120 keV. These fitted resonance parameters (RRR & URR) were used to replace the existing File 2 in the JEFF-3.3 evaluation, without any other changes to the file (e.g., File 1, File 3, etc...). The MCNP transmission calculation using the JEFF-3.3 file with a new RPI evaluation is shown in Fig. 5.17 along with JEFF-3.3 and ENDF/B-VIII.0.
Figure 5.17: The grouped thick-sample transmission measurement in red, along with the MCNP calculated transmission using various evaluated cross sections. The JEFF-3.3 evaluation was modified to include RRR and URR parameters from the analysis described in §5.1 and §5.2, respectively. The new transmission curve using the modified JEFF-3.3 file was plotted in black.

It should be noted that changing from the JEFF-3.3 value of $R' = 7.8$ fermi to $R' = 7.85$ fermi in the URR is important to improve the transmission discrepancy in the low energy URR back towards the measured transmission in the new RPI modified evaluation as compared to the JEFF-3.3 evaluation. Using average level spacing based on the RRR measured $D = 4.17$ eV also reduces the transmission in the URR towards measured data. Using the new resonance parameters in the RRR improves the MCNP calculation in the RRR to match the measured data more closely as seen from 200→2500 eV.

5.3.3.2 Extension of the RRR

It has been shown that in the region from 330→2500 eV that the extension of the RRR better models the transmission measurement, even though levels were being missed. Taking this logic further, the next question would be how far can the RRR be extended before it is no longer the best representation of the cross section? An example can be demonstrated by extending the RRR to 4 keV for $^{181}$Ta and comparing it to the thick-sample transmission measurement. In Mughabghab’s Atlas of Neutron Resonance Parameters [55] $^{181}$Ta had
resolved resonance parameters listed up to 4 keV. The $\Gamma_n$ parameters from Mughabghab were used as the prior input parameters for SAMMY from 2.5→4 keV. Mughabghab only lists sparse $\Gamma_\gamma$ parameters, so an average $\Gamma_\gamma$ value was supplied to SAMMY as the prior input $\Gamma_\gamma$ parameter for resonances from 2.5→4 keV. The spin values of the (s-wave) resonances were randomly selected for each resonance with a $g_\lambda = \frac{2\cdot 3.0 + 1}{2(2\cdot 3.5 + 1)} = 43.75\%$ probability for $J = 3.0$, and a $g_\lambda = \frac{2\cdot 4.0 + 1}{2(2\cdot 3.5 + 1)} = 56.25\%$ probability for $J = 4.0$. The prior parameters were then fit using the evaluation datasets for transmission and capture yield. The new fit is shown for a selected region near 4 keV in Fig. 5.18.

![Figure 5.18: The measured transmission and capture yield data from the RPI measurement. The fit here had been extended to 4 keV using Mughabghab’s listed parameters in the Atlas [55]. By extending the fit to 4 keV, the RRR model was being applied to a region where the measurement was not completely resolved. Despite this, the RRR model recreated the measured transmission and capture yield reasonably well.](image)

Extending the RRR to 4 keV was a demonstration of pushing the RRR upper boundary to well beyond the energy at which resonances are being missed. In Fig. 5.18 it can be seen that the fit matched reasonably well in the capture yield and transmission measurements. It can also be seen that resonances were not completely resolved. If too many resonances were being missed it’s possible that broadening effects such as resolution or Doppler broadening
would not have been properly accounted for. The newly fitted resonance parameters for resonances from 4 to 4000 eV were used to model cross section and tested against the thick-sample transmission measurement. This is shown in Fig. 5.19.

![Graph showing transmission measurement and model predictions](image)

Figure 5.19: The thick-sample transmission measurement and evaluated transmissions are compared to the RPI extended RRR evaluation in black. The extension of the RRR caused some discrepancies between 3 and 4 keV, and no longer seemed to follow the grouped structure of the experimental data. The additional resonances from 2.5 to 4 keV also contribute enough cross section interference to reduce the potential scattering (and increase the transmission) at 2 keV.

The agreement of the calculation with the measured data became worse as the RRR was extended past 3.5 keV to approximately 4 keV. This was likely due to missing resonances. It can also be theorized that the additional resonances from 2.5 to 4 keV negatively interfere with the potential scattering close to 2 keV, creating the discrepancy with the experimental data at 2 keV.

### 5.3.3.3 Multiple Fits of the URR

It’s possible that one set of average resonance parameters would not sufficiently describe the entire URR. In that case, it may be beneficial to fit separate energy regions with different
sets of average parameters. The SAMMY 8.1 FITACS implementation had this capability built in; it would fit multiple energy regions with separate $\tilde{S}_l$, $R_l^\infty$, $\langle \Gamma_{l,\gamma} \rangle$, and $D$.

The SESH program, however, only allowed input for one set of resonance parameters which are used to calculate simulated observables (transmission and capture yield) and theoretical cross sections. This adds additional complexity to the iterative fitting process described in Fig. 5.10 of §5.2. For this reason, a Python [86] program was developed for the present work (by Brown) to be used as a function within the “nuctools” package. The function, called as “sesh_fitacs()”, was used to automate the input and output of the SESH and FITACS programs and test the convergence of the correction factor in each iteration. By writing a program to iterate over the fitting and correction process, less mistakes are likely to be made and the analysis is more reproducible, in addition to being more efficient.

The 1, 3, and 6 mm average total cross section and 1 and 2 mm capture cross section datasets are fit using separate sets of resonance parameters in the energy regions: $2 \rightarrow 10$ keV, $10 \rightarrow 45$ keV, and $45 \rightarrow 120$ keV. Using the SESH and FITACS Python convergence program, upon the $2^{nd}$ iteration the correction factor calculated by SESH to correct the data was < 1% different from the $1^{st}$ iteration. The fitted total and capture cross sections and measured data are shown in Fig. 5.20 and Fig. 5.21.
Figure 5.20: The average total cross section for the 3 and 6 mm Ta measurements, along with the fitted theoretical total cross section in red. The fitted cross section comes from average resonance parameters fit in the energy regions $2 \rightarrow 10$ keV, $10 \rightarrow 45$ keV, and $45 \rightarrow 120$ keV.
Figure 5.21: The average capture cross section for the 1 and 2 mm Ta capture measurements, along with the fitted theoretical capture cross section in red. The fitted cross section comes from average resonance parameters fit in the energy regions $2 \rightarrow 10$ keV, $10 \rightarrow 45$ keV, and $45 \rightarrow 120$ keV.

The jump in theoretical total and capture cross sections in Figs. 5.20 and 5.21 at 45 keV is a result of the different fitted energy regions. This break point was chosen intentionally to assess whether data, that seemed to display different energy dependent behavior in different energy regions, could be modeled better with an entirely different set of parameters. The resulting simulated transmission using the multi-region fit in the URR is shown in Fig. 5.22. Using multiple energy regions to fit the experimental data did not have any pronounced improvement on matching the validation transmission measurement as compared to the full region fit. For this reason it was assumed that a single set of parameters were sufficient for fitting the URR of $^{181}$Ta from 2.4 to 100 keV.
Figure 5.22: The validation transmission measurement and evaluated transmissions compared the RPI evaluation with a multiple energy region fit of the URR. Fitting multiple energy regions in the URR was not seen to have a pronounced improvement over fitting the full energy range of experimental data.

5.3.4 Effect on Criticality

As discussed in Chapter 1 integral benchmarks are an important part of nuclear data evaluation. Integral benchmarks which measure criticality serve to validate the evaluated cross section with a single experimental value: the multiplication factor $k_{eff}$. It’s important to use integral and differential benchmarks together to evaluate cross sections. Integral benchmarks provide a very accurate experimental value that can test the overall quality of evaluated cross sections, and differential benchmarks are capable of identifying more subtle inaccuracies in the cross section. Integral benchmarks depend on the accuracy of the weighted sum of all the cross sections for all materials in the system. This means that if, e.g., the capture cross section for one isotope is over-predicted and the capture cross section for another isotope is under-predicted, they may compensate for each other in a simulated benchmark and still match the experimental $k_{eff}$. To verify isotope specific reaction cross sections, differential data should be consulted. To observe changes in $k_{eff}$ resulting from changes in the upper RRR and lower URR $^{181}$Ta cross section, a benchmark sensitive to cross section in these energy regions must be consulted.
In 2014 it was recognized in a report by Percher et. al. [25] that there was a criticality safety need for more epithermal and intermediate energy criticality benchmarks. Following that motivation, a set of Thermal/Epithermal eXperiments (TEX) were designed to create intermediate energy criticality benchmarks. Tantalum was chosen to be the first material investigated with the TEX benchmarks, which provided an opportunity to test the new evaluation presented in this work, and compare the conclusions drawn from the TEX results and the thick-sample transmission measurement. The TEX MCNP model obtained for this work [87] was preliminary. This means that the model did not yet contain the exact dimensions and geometry of the experiment, and the exact value of $k_{eff}$ was not known. The information that could be drawn from the TEX benchmarks was how impactful the Ta evaluation was to the criticality of the system.

More information about the TEX benchmarks can be found in Ref. [25], but in general the TEX benchmarks consisted of plutonium fuel moderated by polyethylene and a diluent material of interest (in this case Ta) which was distributed throughout the assembly. There were multiple experimental designs with differing amounts of polyethylene to vary the moderation of the neutrons. The design shown in this work was the design with the greatest sensitivity to the energy range of interest ($\approx 0.2 \rightarrow 100$ keV): “Experiment 8”. Experiment 8 was modeled in MCNP with all ENDF/B-VIII.0 isotopes, and only the $^{181}$Ta evaluation was swapped to observe the impact of the Ta cross section representation. The computed $k_{eff}$ values $C$, for the ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 libraries are normalized to the computed $k_{eff}$ using the ENDF/B-VIII.0 evaluation of Ta: $C_{ENDF}$, and plotted together in Fig. 5.23.
Figure 5.23: The computed $k_{\text{eff}}$ values $C$ corresponding to the simulation of the TEX [25] benchmarks, with Ta cross sections reported in the ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 libraries, normalized to the computed $k_{\text{eff}}$ value using the ENDF/B-VIII.0 evaluation of Ta: $C_{\text{ENDF}}$. This comparison shows the impact of using the reported cross sections for Ta from different evaluated libraries. The difference in $k_{\text{eff}}$ between JENDL-4.0 and ENDF/B-VIII.0 is $\Delta k \approx 800$ pcm or 0.8%.

It’s obvious from the normalized computed $k_{\text{eff}}$ values in Fig. 5.23 that the discrepancies between the ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 Ta cross sections have a significant impact on the expected $k_{\text{eff}}$: up to $\Delta k \approx 800$ pcm (0.8%). In this context $\Delta k$ is defined as seen in Eq. 5.11, where the $k_{\text{eff}}$ values are computed by MCNP.

$$\left( \frac{\Delta k}{k} \right)_{\text{JEFF}} = \frac{k_{\text{eff,JEFF}} - k_{\text{eff,ENDF}}}{k_{\text{eff,ENDF}}}$$  \hspace{1cm} (5.11)

The new preliminary RPI evaluation can also be tested with the TEX benchmark. The $C/C_{\text{ENDF}}$ values corresponding to the RPI evaluation are shown in Fig. 5.24. The $C/C_{\text{ENDF}}$ value marked as “McD. URR” comes from modeling the $^{181}\text{Ta}$ cross section with a modified JEFF-3.3 evaluation where only the URR average values have been changed to the McDermott evaluated values. The “only URR” evaluation is similar, it is the $C/C_{\text{ENDF}}$. 
where only the RPI evaluated URR parameters have replaced parameters in the JEFF-3.3 file. The purpose of the “McD. URR” and “only URR” evaluation is to show the impact of the small difference in cross section between the JEFF-3.3 and RPI evaluations in the URR: a change of $\Delta k \approx 200$ pcm from JEFF-3.3 in the TEX benchmark. Using the full preliminary RPI RRR/URR evaluated parameters to modify the JEFF-3.3 evaluation results in a change of $\Delta k \approx 700$ pcm from JEFF-3.3.

Figure 5.24: The computed $k_{eff}$ values corresponding to the simulation of the TEX [25] benchmarks, with Ta cross sections reported in the ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 libraries (squares) $C$, normalized to the computed $k_{eff}$ value using the ENDF/B-VIII.0 evaluation of Ta, $C_{ENDF}$. Also shown are the $C/C_{ENDF}$ values for the URR and full RRR/URR RPI evaluation (circles). The difference in $k_{eff}$ between JEFF-3.3 and the RPI “URR only” evaluation is approximately $\Delta k \approx 200$ pcm. This difference highlights the importance of the URR cross section and resonance self shielding effect for criticality.

The fact that the full RPI evaluated parameters resulted in a calculated $k_{eff}$ very similar to that of ENDF/B-VIII.0 could be misleading (as we don’t have an experimental

---

2Preliminary as it is constrained by the JEFF-3.3 thermal cross section during the evaluation process. RPI evaluation of the RRR cross section, in particular low energy RRR, is on-going.
Table 5.3: The capture resonance integrals ($I_\gamma$) for the various evaluations as calculated by NJOY21 [62]. The ENDF/B-VIII.0 value is greatest, indicating that it will have the greatest neutron absorption. Also note the proximity of the RPI and ENDF/B-VIII.0 values as compared to that of JEFF-3.3. This is consistent with the TEX benchmark results presented in Fig. 5.24.

<table>
<thead>
<tr>
<th>Library</th>
<th>$I_\gamma$ [b]</th>
</tr>
</thead>
<tbody>
<tr>
<td>ENDF/B-VIII.0</td>
<td>739</td>
</tr>
<tr>
<td>RPI</td>
<td>719</td>
</tr>
<tr>
<td>JEFF-3.3</td>
<td>660</td>
</tr>
</tbody>
</table>

$k_{eff}$, but also served to make an important point: the weighted integration of ENDF/B-VIII.0 $^{181}\text{Ta}$ cross section may have been correct even while the point wise cross section was very obviously wrong over certain energy regions. For example, the capture resonance integral $I_\gamma$ for the RPI evaluation, ENDF/B-VIII.0, and JEFF-3.3 are shown in Table 5.3. This reinforces the importance of validating evaluated cross section with both integral and differential data.

Beyond a simple resonance integral one can further ascertain the impact evaluated cross sections have on the computed $k_{eff}$ value by calculating the sensitivity ($\frac{\Delta k_{eff}}{k_{eff}} / \frac{\Delta \sigma_x}{\sigma_x}$) for $k_{eff}$ as a function of energy. The MCNP program includes functionality to determine the sensitivity of the calculated $k_{eff}$ value to the input cross sections. This sensitivity can be determined as a function of energy, in order to better understand how $k_{eff}$ is affected by cross section in different energy regions. The sensitivity of the computed $k_{eff}$ to the RPI evaluated capture ($\sigma_\gamma$) and elastic scattering ($\sigma_n$) cross sections is shown in Fig. 5.25.
Figure 5.25: The sensitivity ($\frac{\Delta k_{eff}/k_{eff}}{\Delta \sigma_n/\sigma_n}$) of the computed $k_{eff}$ value to the RPI evaluated capture ($\sigma_\gamma$) and elastic scattering ($\sigma_t$) cross sections. The TEX benchmark is more sensitive to the capture cross section than the scattering cross section. It should be noted that $\sigma_t \approx \sigma_\gamma + \sigma_n$ for energies $< 100$ keV.

We can see from Fig. 5.25 that the $k_{eff}$ value is much more sensitive to the capture cross section than the elastic scattering cross section. This is why the ENDF value and RPI value for $k_{eff}$ are comparable: because they have similar capture cross sections.

The combination of the integral and differential benchmarks presented herein (TEX and thick-sample Ta transmission) provide clarity on the inaccuracies that may exist in a given cross section evaluation. They make it possible to identify selected portions of the cross section and their corresponding impact on the computed $k_{eff}$. The thick Ta transmission presented in this work (Brown) extends this capability in the URR, specifically for identifying weaknesses in average resonance parameters and the resonance self-shielding correction in continuous energy Monte Carlo codes.
CHAPTER 6
CONCLUSIONS

The focus of this Ph.D. work has been in two areas: taking measurements that can be used to perform a new evaluation of the isotope $^{181}$Ta and creating a self-shielding URR transmission validation method. The high-resolution measurements presented herein have increased the quality of RRR and URR transmission and capture yield datasets available to the public. The capture yield data presented here had the highest resolution to date of any publicly available dataset and played an important role in constraining the evaluated average URR parameters. The value of the transmission measurements presented here was less about the resolution (as the Harvey data were well resolved) and more about the multiple sample thicknesses, and the thicker sample transmissions in the URR which provide more accurate cross section information from $\approx 3$ to 100 keV than was previously available. The thick-sample transmission validation method has been shown to be a valuable new tool for evaluators to validate the quality of nuclear data evaluations in the URR and upper RRR. Conclusions drawn from these measurements and the analysis that followed them are presented here in Chapter 6.

6.1 $^{181}$Ta Evaluation

In terms of $^{181}$Ta, following the new measurements, we can more definitively draw conclusions on discrepant evaluations. The ENDF/B-VIII.0 library was the most inaccurate in the URR and upper RRR. It should also be noted that ENDF/B-VIII.0 was based on older evaluations and so had not been updated for several decades. The value for the effective radius $R'$ was larger than all other evaluated effective radii and did not match well with the RPI transmission data. The thermal cross sections of ENDF/B-VIII.0 were also inconsistent with other libraries and those published by Mughabghab [55]. ENDF/B-VIII.0 should be re-evaluated and the RRR should be extended to higher energies, possibly to 2.4 keV, in order to improve agreement with measured data.

The URR average parameters in the JEFF-3.3, JENDL-4.0, and ENDF/B-VIII.0 libraries should be re-evaluated as well. The ENDF/B-VIII.0 library would benefit from having the URR extended to 100 keV, so as to correct for resonance self-shielding beyond
5 keV. The JEFF-3.3 and JENDL-4.0 libraries need to have average level spacings which properly account for resonance self-shielding and should update average parameters $\langle \Gamma_n \rangle$ and $\langle \Gamma_\gamma \rangle$.

6.2 Differential Validation Transmission

The energy differential thick-sample transmission measurement has been shown to be capable of validating the URR and upper RRR. This novel application of a thick transmission measurement was valuable as it validated cross section in a historically neglected energy region: the URR. While there have been examples of benchmark transmission in the past, most focused on the high energy region and optical model, instead of the URR where resonance self-shielding plays a significant role in transport calculations.

One of the points of interest that this validation method can illuminate is whether the extension of the RRR into a region where levels are being missed has an overall negative or positive impact on the cross section representation. In general, the validation transmission measurement supported the extension of the RRR past the resolution limit for $^{181}$Ta. This was based on the energy region used in JEFF-3.3: up to 2.5 keV. It can be seen that as the RRR was extended to 4 keV, however, the RRR representation seemed to perform worse than a URR formalism.

This thick-sample transmission measurement has been showcased identifying specific inaccuracies in the cross section, and how much these inaccuracies affect systems of criticality, e.g., the difference of $\frac{\Delta k}{k} \approx 200$ pcm from only changing the URR parameters. The validation transmission method in this work should be used alongside standard integral benchmarks. Integral benchmarks provide a highly accurate measure of the weighted sum of all cross sections, and the thick-sample transmission measurement complements them by identifying weaknesses in specific isotopes and energy regions. The validation transmission measurement presented herein is an accurate new tool that will allow evaluators to increment evaluations closer to the true cross section.

Ultimately the conclusions of the work can be summarized in bulleted points as follows:

- New high resolution measurements of $^{181}$Ta made at RPI allow evaluators to determine more accurate resonance parameters.
A novel use of a thick-sample transmission measurement was developed to validate URR evaluated parameters, further improving the accuracy of future evaluations.

The novel thick-sample transmission measurement was further shown to:

1. Validate the extension of the RRR in $^{181}$Ta up to 2.5 keV
2. Validate the extension of the URR in $^{181}$Ta up to 100 keV
3. Identify inaccuracies introduced by over-extension of the RRR, in this case to 4 keV

Evaluators can use these new measurements and methods to improve the accuracy of evaluated cross sections, ultimately enabling greater accuracy in modeling of nuclear applications.

### 6.3 Future Developments

The research direction that this work leads to has many components, including new evaluation measurements, improvements to the capture detector hardware and software, and new experimental covariance analysis frameworks.

#### 6.3.1 Thermal Data

During the evaluation of the RRR with SAMMY it was found that the thermal cross section of $^{181}$Ta was not very well known. There have been many measurements of the thermal cross section, but the measurements were inconsistent, and some cross sections were published without uncertainty. The thermal value of the cross section is important to the fit along with the RRR as it affects low lying resonances, in this case the 4.3 eV resonance of $^{181}$Ta. This is the reason why the JEFF-3.3 cross section was used to constrain the RRR fit from 0.01 to 0.1 eV for the present work (Brown). The inclusion of JEFF-3.3 thermal data biases the evaluation performed in this work, however. In the future a new thermal cross section measurement should be made, or a thorough analysis of the available thermal data should be performed to understand the inconsistencies found between experimental data. An example of the thermal values for total and capture cross section compiled by Barry [88] from EXFOR [34] are shown in Fig. 6.1 and Fig. 6.2. It should be noted that this is not an exhaustive list but that it is representative of the available data.
Figure 6.1: A compilation of thermal total cross section values compiled by Barry [88]. The horizontal solid and dashed red lines indicate the evaluated value published by Mughabghab in the Atlas of Neutron Resonances [55].

Several of the data in Fig. 6.1 had to be interpolated or extrapolated from the published data; these were plotted without error bars. The horizontal solid and dashed red lines in Figs. 6.1 and 6.2 indicate evaluated thermal cross section values and their estimated uncertainty as published in the Atlas of Neutron Resonances [55].

6.3.2 Upgrade of the Capture Detector

6.3.2.1 Angular Distributions

The capture detection system designed and built by McDermott [33] at the RPI Linac uses an SIS3305 digitizer by Strucke Innovative Systems [89] to read the signal from the four C$_6$D$_6$ detectors. This digitizer was capable of accepting up to 8 inputs, allowing for the possibility of increasing the number of detectors used for the capture detection system. With the addition of 3 or 4 more detectors it may be possible to observe average angular distributions in capture cascades. The detectors would be moved further away from the sample of interest and placed at several different angles with respect to the incident neutron beam (the cascades should be isotropic orthogonal to the neutron beam). The angular distribution of the cascade photons could be used to determine spin assignment for resonances, e.g., if
the distribution is isotropic the quantum angular momentum $L = 0$, if there are anisotropies in the distribution $L > 0$ and the orientation of the distribution depends on the quantum state of the resonance. This process would involve software upgrades to the data acquisition software to recognize the new data inputs. Upgrades to the data post-processing software would also need to include coincidence handling to identify photons originating from the same capture cascade.

### 6.3.2.2 Detector Mounting System

Another improvement to the capture system is a detector mounting system of greater stability and reproducibility. In an effort to reduce the mass of structural materials surrounding the sample, detectors are suspended by an Al rod. The Al rod suspension mounts achieve a specific goal: reducing the probability for neutron captures in structural material that would contribute to the background of the measurement. It is not, however, possible to set detectors at specific distances and angles to the capture sample with accurate reproducibility. To achieve this, a new mounting system should be designed. A simple design that would improve on the original is an Al stand shaped similarly to a music stand. This would
keep most of the mass close to the floor and allow more accurate and reproducible methods of setting the position and angle of detectors.

6.3.2.3 Background Shielding

Thought should also be given to increased shielding of the detectors. It was found by McDermott [50] that the natural room background mostly came from photons originating from the floor and surrounding ground surface. By shielding the detectors with a ring (radius \( \approx 1 \text{ m} \)) of Pb bricks \( \approx 5.08 \text{ cm} \) thick and 50 cm high, a reduction in room background of 1.5-2x is possible. This Pb ring would need to be elevated to the appropriate height so as to project the greatest solid angle on the floor from the perspective of the detectors. An example of shielding geometry modeled in MCNP is shown in Fig. 6.3.

Figure 6.3: Pb shield designed to reduce the room background measured on by the capture system by a factor of \( \approx 1.5-2x \).

Supporting the mass of Pb described is achievable and would be best employed on stands with wheels so that the shielding could be transported easily to other detection systems. Preliminary modeling and analysis show that the majority of Linac beam neutrons that scatter off of a sample towards the Pb ring would not be scattered back. It’s very important to minimize the neutron return from any shielding, and deserves further study to ascertain whether additional scattered neutron background would introduce unwanted signal in a measurement.

6.3.3 Upgrade of the Transmission Detector

The 100 m transmission detector (MELINDA) used a set of analog electronics which received voltage pulses, discriminated them (with LLD and ULD) and summed the four mod-
ules to be recorded as a single detector as a function of time. It may be beneficial to read the
detectors with a digital system. This would provide greater freedom to the experimentalist
in how to discriminate voltage pulse information. If a CLYC detector were incorporated
into the detection system pulse shape discrimination could be achievable [90][91], and back-
ground characterization analysis could be performed. The neutron and photon background
measured by the 100 m detector is substantial in the mid-keV energy region and remains
an existing problem for the MELINDA detector system. This includes a significant time-
dependent room background measured outside of the Linac beam, meaning there is a room
return of photon and/or neutrons. Several preliminary measurements have been made to try
to identify this background, where the evidence suggests that it is likely in-beam photons.
BIBLIOGRAPHY


[36] V. Konks, Y. P. Popov, and F. Shapiro, “Cross sections for radiative capture of neutrons with energies up to 50 kev by \(^{139}\)La, \(^{141}\)Pr, \(^{181}\)Ta, and \(^{197}\)Au nuclei,” Sov. Phys. JETP, vol. 19, no. 1, pp. 59–64, Jul. 1964.


[68] S. Wang et al., “The RPI multiplicity detector response to γ-ray cascades following neutron capture in $^{149}$Sm and $^{150}$Sm,” Nuc. Instr. and Meth. in Phys. Research


A.1 Fortran Program: Sampling the Wigner Distribution

```fortran
program sample_distribution

  implicit none

  ! ---- Declare variables -----------------------------------------------
  ! ----
  integer(kind=4) :: i
  real :: D ! average level spacing
  real :: random ! random number
  real :: pi
  integer :: x_loc ! where to bin
  integer, dimension(60) :: bins ! bin structure
  real, dimension(60) :: xaxis ! axis to

  ! -------------------------------------------------------------------------
  ! ---- Set some variables -----------------------------------------------
  bins = 0 ! all bins start at zero
  pi = 3.14159
  D = 0

  ! ------------------------------------------------------------------------
  ! ---- Run a Monte Carlo sampling of Wigner distribution -----------------
  do i=0,100000000
    random = rand(0)
    call wigner(D,pi,random) ! set the normalized D/<D>
    x_loc = nint(D*10)+1 ! where to bin the D/<D>
    bins(x_loc) = bins(x_loc) + 1 ! increment bin by 1
  end do
```
! ---- Print out the binned widths (D/<D>) --------------------------------

do i=1,60
   xaxis(i) = dble(i)/10.0
   print*, xaxis(i), bins(i)
end do

end program sample_distribution

subroutine wigner(D,pi,random)
   implicit none
   real :: D
   real :: random
   real :: pi

   D = 2/sqrt(pi)*sqrt(-log(random))

end subroutine wigner

A.2 Transmission Benchmark MCNP Input File

C ------- End pipe scattering background MCNP input file -------
C ------- Cell Definitions -------
C ------- Pipe window -------
C
c Aluminum cell
1 1 -2.70 -1 IMP:N=1
C -------------------
c Det cell
2 0 -2 IMP:N=1
C -------------------
c Tantalum sample
3 3 -16.92 -3 IMP:N=1
C -------------------
c ----- Control Volume ----- 
c 5 2 -0.001205 -5 #1 #2 #3 #4 IMP:N=1
4 0 -4 #1 #2 #3 IMP:N=1
5 0 4 IMP:N=0

C ------------------------------------------------------------------------
c Surface Definitions |
C ------------------------------------------------------------------------
c --- Pipe window --- 
c Aluminum 
c --- base at (0,0,100), top at (0,0,1/32inch), R=45.72 (1.5 ft)
1 RCC 0 0 100 0 0 0.079375 45.72
C ------------------------------------------------------------------------
c --- Detector --- (xmin, xmax, ymin, ymax, zmin, zmax)
c --- x,y = 16x16 inches, z = 0.5 inch,
c --- detector surface facing beam centered at origin
2 RPP -20.32 20.32 -20.32 20.32 -1.27 0
C ------------------------------------------------------------------------
c Sample (xmin, xmax, ymin, ymax, zmin, zmax)
c (width, length, thickness)
3 RPP -5.219 5.219 -5.142 5.142 1998.8 2000
C ------------------------------------------------------------------------
c ---- World boundary
4 SO 10100

C Data Cards
C --------------------------------
C Source Definition
C --------------------------------
mode n
sdef pos=0.0 0.0 3518 dir=-1 vec=0 0 1 ara=3.14 tme=D9 erg=D1
    axs=0.0 0.0 1.0 rad=D10 par n
c ============================ FLUX from 5 eV to 450 keV, 102 bins ======
    si1 A 5.6526E-05 6.1483E-05 6.6874E-05 7.2738E-05 7.9117E-05 8.6054E-05
        9.3600E-05 1.0181E-04 1.1074E-04 1.2045E-04 1.3101E-04 1.4250E-04
        1.5499E-04 1.6858E-04 1.8337E-04 1.9945E-04 2.1693E-04 2.3596E-04
        1.1653E-03 1.2675E-03 1.3786E-03 1.4995E-03 1.6310E-03 1.7740E-03
        1.9296E-03 2.0988E-03 2.2828E-03 2.4830E-03 2.7007E-03 2.9376E-03
        3.1952E-03 3.4754E-03 3.7801E-03 4.1116E-03 4.4721E-03 4.8643E-03
        5.2908E-03 5.7548E-03 6.2594E-03 6.8083E-03 7.4054E-03 8.0547E-03
        8.7611E-03 9.5293E-03 1.0365E-02 1.1274E-02 1.2262E-02 1.3338E-02
        1.4507E-02 1.5779E-02 1.7163E-02 1.8668E-02 2.0305E-02 2.2086E-02
        2.4022E-02 2.6129E-02 2.8420E-02 3.0912E-02 3.3623E-02 3.6572E-02
        3.9779E-02 4.3267E-02 4.7061E-02 5.1188E-02 5.5676E-02 6.0558E-02
        6.5869E-02 7.1645E-02 7.7927E-02 8.4761E-02 9.2194E-02 1.0028E-01
        1.0907E-01 1.1864E-01 1.2904E-01 1.4035E-01 1.5266E-01 1.6605E-01
        2.0000E-01 2.5000E-01 3.0000E-01 3.5000E-01 4.0000E-01 4.5000E-01
    sp1 1.60194E-05 1.64435E-05 1.63429E-05 1.68205E-05 1.61953E-05 1.63213E-05
        1.65849E-05 1.63864E-05 1.70546E-05 1.73831E-05 1.77910E-05 1.75672E-05
        1.78444E-05 1.89543E-05 1.79260E-05 1.88409E-05 1.83507E-05 1.81549E-05

si9 0.0 1.0 $ 10 ns pulse width
sp9 0.0 1.0 $ Distributed uniformly
si10 3.0 $ 3 cm radius beam
sp10 -21 1 $ uniform flux

NPS 1e8

C

C Tallies

F11:N 2.5

C (Default electron and photon cutoff energies are 1 keV)

C Tally time bin structure (shakes) (~1e6 eV)

T11 350 2046i 5e4

FQ0 T U

C
Material Definitions

C -- Aluminium 6061 --
M1 12024 0.011162 13027 0.977325 14028 0.005796 &
   22048 0.000499 24052 0.001017 25055 0.000435 &
   26056 0.001987 29063 0.001174 30064 0.000606

C -- Air --
M2 6012 -0.000124 7014 -0.755268 8016 -0.231781 18038 -0.012827

C -- Tantalum -- (99c here should be the RPI urr mod for JEFF 3.3)
M3 73181 1 NLIB=95c

C -- Na --
C M3 11023 1.0

C -- C6D6 --
C

C M2 6012 -0.856390 1002 -0.1436

C -- Silicone Rubber --
C M3 1001 0.597039 6000 0.199359 8016 0.104169 &
   14000 0.099434

C -- Quartz --
C M4 8016 0.666667 14000 0.333333

C -- Copper --
C M5 29000 1

C -- Mu Metal --
A.3 FITACS Input File

This is the FITACS Input file for fitting 3 total cross section data sets and 2 capture cross section data sets with 3 sets of resonance parameters in different energy regions.

Ta-181 urr par file

 ITERATIONS.= 2
 TOLERANCE. = 0.000005
 RADIUS = 7.800
 AW. = 180.947996

ELASTIC AND INELASTIC STATES

 0.0  3.5  1.0
6237.0  4.5  -1.0
136262.0  4.5   1.0  
158554.0  5.5  -1.0  
301622.0  5.5   1.0  
337540.0  6.5  -1.0  
482168.0  2.5   1.0  
495184.0  6.5   1.0  
542510.0  7.5  -1.0  
590060.0  3.5   1.0  
615190.0  0.5   1.0  
618990.0  1.5   1.0  
716659.0  7.5   1.0  
772970.0  8.5  -1.0  
892900.0  5.5   1.0  
965000.0  8.5   1.0  
994200.0  2.5  -1.0  
1022600.0  4.5  -1.0  
1028000.0  9.5  -1.0  
1085600.0  6.5   1.0  
1163600.0  6.5  -1.0  
1205700.0  1.5   1.0  
1239470.0  9.5   1.0  
1278100.0  2.5   1.0  
1304800.0  7.5   1.0  
1307110.0 10.5  -1.0  

--------------------------  
BINDING ENERGY (in MeV) = 7.57680000  
PAIRING ENERGY (in MeV) = 0.73000000  
--------------------------

STRENGTH  DEL_S  DISTANT  DEL_D  GAM_WIDTH  DEL_G  BETHED  
  0.000185  0.0000110  -0.006600  0.0010000  0.0678000  0.0110000  4.1700000
MINIMUM ENERGY in eV = 2000.0
ENERGY MAXIMUM in eV = 10000.0

BINDING ENERGY (in MeV) = 7.57680000
PAIRING ENERGY (in MeV) = 0.73000000

STRENGTH  DEL_S   DISTANT  DEL_D  GAM_WIDTH  DEL_G  BETHED
0.000185  0.0000110 -0.006600  0.0010000  0.0678000  0.0110000  4.1700000
0.000050  0.0000200  0.0000000  0.0100000  0.0678000  0.0110000
0.000230  0.0000300  0.0000000  0.0100000  0.0678000  0.0110000

ENERGY MAXIMUM in MeV = 0.045

BINDING ENERGY (in MeV) = 7.57680000
PAIRING ENERGY (in MeV) = 0.73000000

STRENGTH  DEL_S   DISTANT  DEL_D  GAM_WIDTH  DEL_G  BETHED
0.000185  0.0000110 -0.006600  0.0010000  0.0678000  0.0110000  4.1700000
0.000050  0.0000200  0.0000000  0.0100000  0.0678000  0.0110000
0.000230  0.0000300  0.0000000  0.0100000  0.0678000  0.0110000

ENERGY MAXIMUM in MeV = 0.120
END OF RESONANCE PARAMETER DESCRIPTION

-------------------------------
NORMALIZATIONS

TOTAL  1.000000  0.000000  0.000000  0.000000  0.000000  0.000000
TOTAL  1.000000  0.000000  0.000000  0.000000  0.000000  0.000000
TOTAL  1.000000  0.000000  0.000000  0.000000  0.000000  0.000000
CAPTURE  1.000000  0.000000  0.000000  0.000000  0.000000  0.000000
CAPTURE  1.000000  0.000000  0.000000  0.000000  0.000000  0.000000

A.4 SESH Input File

Please note that the SESH input requires that 36 lines are present in the input file. Add return characters (new lines) after the energy and iteration specifications are complete to meet the requirement. Greater detail on the input file is given in the report by Fröhner [63].

2mm Ta-181 Multiple Scattering & Self Shielding Correction 0
181.0 1.0 7.57680 0.730 294. 3.50
0.06291 4.17000 1.575e-04 0.00000 7.80000 1.00000
0.06928 4.17000 7.308e-05 0.00000 7.80000 1.00000
0.06291 4.17000 2.303e-04 0.00000 7.80000 1.00000
5.660E-03 1.713E-02 3.358E-02 5.631E-03 1.115E-02
0.000E+00 0.000E+00 0.000E+00 3.351E-01 3.351E-01
0.000E+00 0.000E+00 0.000E+00 0.000E+00 0.000E+00
1.377E-01 1.377E-01 1.377E-01 1.377E-01 1.377E-01
17.
1.50000 15000. 1.75000 15000. 2.00000 15000. 2.25000 15000.
2.50000 15000. 2.75000 15000. 3.00000 15000. 3.25000 15000.
3.50000 15000. 4.00000 15000. 4.50000 15000. 5.00000 15000.
5.50000 15000. 6.00000 15000. 6.50000 15000. 7.00000 15000.
7.50000 15000. 8.00000 15000. 8.50000 15000. 9.00000 15000.
| 9.50000 | 15000. | 10.0000 | 15000. | 10.5000 | 15000. | 11.0000 | 15000. |
| 11.5000 | 15000. | 12.0000 | 15000. | 12.5000 | 15000. | 13.0000 | 15000. |
| 14.0000 | 15000. | 15.0000 | 15000. | 16.0000 | 15000. | 17.0000 | 15000. |
| 18.0000 | 15000. | 19.0000 | 15000. | 20.0000 | 15000. | 21.0000 | 15000. |
| 22.0000 | 15000. | 23.0000 | 15000. | 24.0000 | 15000. | 25.0000 | 15000. |
| 26.0000 | 15000. | 28.0000 | 15000. | 30.0000 | 15000. | 32.0000 | 15000. |
| 36.0000 | 15000. | 40.0000 | 15000. | 44.0000 | 15000. | 48.0000 | 15000. |
| 52.0000 | 15000. | 56.0000 | 15000. | 60.0000 | 15000. | 70.0000 | 15000. |
| 80.0000 | 15000. | 90.0000 | 15000. | 100.000 | 15000. | 110.000 | 15000. |
| 120.000 | 15000. | 130.000 | 15000. | 140.000 | 15000. | 150.000 | 15000. |
Sample Characteristics

Prior to this Ph.D. work, a 2.02 mm thick 10x10 cm sample of Ta of high purity was used in McDermott’s work [50] and re-used for the present Ph.D.; sample characteristics are listed in Ref [50]. As measurements of multiple sample thicknesses were needed, additional samples were ordered. Two 1 mm samples, one 3 mm sample, and two 6 mm samples were ordered from Stanford Advanced Materials. All of the samples were derived from the same lot (LOT NUMBER: WT170623-5443), and were 99.95% pure elemental Ta. To distinguish each of the 1 and 6 mm samples from each other, a label of “A” and “B” was given to the samples, and written on the face of the samples using a permanent marker. The impurities of the samples are given in Table B.1 in units of parts per million (ppm).

Table B.1: The sample impurities for all the samples obtained in an order from Stanford Advanced Materials, LOT NUMBER: WT170623-5443.

<table>
<thead>
<tr>
<th>Element</th>
<th>PPM</th>
<th>Element</th>
<th>PPM</th>
</tr>
</thead>
<tbody>
<tr>
<td>W</td>
<td>20</td>
<td>Nb</td>
<td>30</td>
</tr>
<tr>
<td>Mo</td>
<td>20</td>
<td>Ti</td>
<td>&lt;1</td>
</tr>
<tr>
<td>O</td>
<td>110</td>
<td>Al</td>
<td>&lt;1</td>
</tr>
<tr>
<td>N</td>
<td>20</td>
<td>Mn</td>
<td>&lt;1</td>
</tr>
<tr>
<td>C</td>
<td>20</td>
<td>Cu</td>
<td>&lt;1</td>
</tr>
<tr>
<td>H</td>
<td>8</td>
<td>Sn</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Fe</td>
<td>20</td>
<td>Ca</td>
<td>&lt;5</td>
</tr>
<tr>
<td>Cr</td>
<td>3</td>
<td>Mg</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Ni</td>
<td>3</td>
<td>Zr</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Si</td>
<td>10</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

In Table B.2 the Ta sample dimensions are listed for the samples used in this work. The dimensions of length, width, and thickness are measured with calipers at 5 different locations along the sample and the average dimensions are listed along with the standard deviation. The mass of the 1 mm samples are measured with a more sensitive scale than the other samples, the mass was measured 5 times and the average mass was taken with standard deviation as uncertainty; for the heavier samples the mass did not vary between weighings. The average density of all the samples is \( \approx 16.92 \text{ g/cm}^3 \). The two 6 mm samples
are stacked together for the thick 12 mm sample transmission benchmark. The areal density and percent uncertainty in the 12 mm stacked sample is $0.06716614 \pm 0.42\%$ at/b.

Table B.2: The sample dimensions and mass are listed for each of the ordered samples. In an effort to be explicit the calculated quantities of density and areal density of the samples are listed as well.

<table>
<thead>
<tr>
<th>Dimension</th>
<th>1 mm A</th>
<th>1 mm B</th>
<th>3 mm</th>
<th>6 mm A</th>
<th>6 mm B</th>
</tr>
</thead>
<tbody>
<tr>
<td>Length [cm]</td>
<td>10.093±0.007</td>
<td>10.050±0.001</td>
<td>10.032±0.002</td>
<td>10.034±0.001</td>
<td>10.022±0.001</td>
</tr>
<tr>
<td>Width [cm]</td>
<td>10.112±0.002</td>
<td>10.055±0.004</td>
<td>10.0208±0.0004</td>
<td>10.0300±0.0008</td>
<td>10.030±0.001</td>
</tr>
<tr>
<td>Thickness [cm]</td>
<td>0.098±0.001</td>
<td>0.0992±0.0008</td>
<td>0.3048±0.0019</td>
<td>0.6064±0.0013</td>
<td>0.6042±0.0021</td>
</tr>
<tr>
<td>Mass [g]</td>
<td>173.7117±0.0004</td>
<td>170.9692±0.0004</td>
<td>517.5±0.1</td>
<td>1015.0±0.1</td>
<td>1015.0±0.1</td>
</tr>
<tr>
<td>$\rho$ [g/cm$^3$]</td>
<td>17.33±0.23</td>
<td>17.06±0.14</td>
<td>16.89±0.11</td>
<td>16.63±0.04</td>
<td>16.71±0.06</td>
</tr>
<tr>
<td>$n$ [at/b]</td>
<td>0.00566±0.00007</td>
<td>0.00563±0.00005</td>
<td>0.0171±0.0001</td>
<td>0.03356±0.00007</td>
<td>0.03360±0.00012</td>
</tr>
</tbody>
</table>
APPENDIX C
Useful Derivations

C.1 $\frac{\partial E}{\partial t}$ for Relativistic Kinetic Energy

The relativistic kinetic energy equation is given by Eq. C.1, where $m_n$ is the neutron mass, $c$ is the speed of light, $FP$ is the length of neutron travel, and the difference of an initial time $t_0$ and time at which a neutron completed traveling the FP $t_i$ is the time of flight of the neutron.

$$E = m_n c^2 \left( \frac{1}{\sqrt{1 - \left(\frac{FP}{t_0} - t_0\right)^2}} - 1 \right) \quad (C.1)$$

For the sake of simplicity, $FP = L$, and $t_i - t_0 = t$. With some manipulation we come to Eq. C.2

$$E = m_n c^2 \left( \left(1 - \frac{L^2}{t^2 c^2}\right)^{-1/2} - 1 \right) \quad (C.2)$$

The derivative of the relativistic kinetic energy equation with respect to time is given by Eq. C.3.

$$\frac{\partial E}{\partial t} = \frac{\partial}{\partial t} \left[ m_n c^2 \left( \left(1 - \frac{L^2}{t^2 c^2}\right)^{-1/2} - 1 \right) \right] \quad (C.3)$$

Multiplying the derivative through, and bringing the constants outside of the derivative results in Eq. C.4.

$$\frac{\partial E}{\partial t} = m_n c^2 \left( \frac{\partial}{\partial u} \left(1 - \frac{L^2}{t^2 c^2}\right)^{-1/2} \right) \quad (C.4)$$

Making a $u$ substitution simplifies the equation as shown in Eq. C.5.

$$\frac{\partial E}{\partial t} = m_n c^2 \left( \frac{\partial}{\partial u} \left( u^{-1/2} \right) \right), \quad u = 1 - \frac{L^2}{t^2 c^2} \quad (C.5)$$

Taking the derivative with respect to $u$ and then reverting to $1 - \frac{L^2}{t^2 c^2}$ gives us Eq. C.6.
\[
\frac{\partial E}{\partial t} = m_n c^2 \left( \frac{-1}{2} \left(1 - \frac{L^2}{t^2 c^2} \right)^{-3/2} \frac{\partial}{\partial t} \left(1 - \frac{L^2}{t^2 c^2} \right) \right) \tag{C.6}
\]

Finally we come to the solution in Eq. C.7.

\[
\frac{\partial E}{\partial t} = m_n c^2 \left( \frac{-1}{2} \left(1 - \frac{L^2}{t^2 c^2} \right)^{-3/2} \left(2 \frac{L^2}{t^3 c^2} \right) \right) \tag{C.7}
\]

This is simplified in Eq. C.8.

\[
\frac{\partial E}{\partial t} = -m_n c^2 \left( \frac{\left( \frac{L^2}{t^3 c^2} \right)}{\left(1 - \frac{L^2}{t^2 c^2} \right)^{3/2}} \right) \tag{C.8}
\]

### C.2 \( \frac{\partial t}{\partial E} \) for Relativistic Kinetic Energy

The equation for the relativistic relation of time of flight (TOF) to kinetic energy \( E \) is given in C.9 with flight path \( FP \), \( c \) as the speed of light, and \( m_n \) as the neutron mass.

\[
TOF = \frac{FP}{c} \frac{1}{\sqrt{1 - \frac{1}{\left( \frac{E}{m_n c^2} + 1 \right)^2}}} \tag{C.9}
\]

For simplicity we will set \( t = TOF \) and \( FP = L \) to begin the derivation: this is shown in Eq. C.10.

\[
t = \frac{L}{c} \frac{1}{\sqrt{1 - \frac{1}{\left( \frac{E}{m_n c^2} + 1 \right)^2}}} \tag{C.10}
\]

The derivative of the \( t \) with respect to \( E \) is given in Eq. C.11.

\[
\frac{\partial t}{\partial E} = \frac{\partial}{\partial E} \left[ \frac{L}{c} \frac{1}{\sqrt{1 - \frac{1}{\left( \frac{E}{m_n c^2} + 1 \right)^2}}} \right] \tag{C.11}
\]

The flight path length \( L \) and speed of light \( c \) are constant, and come out of the derivative for Eq. C.12.
\[
\frac{\partial t}{\partial E} = \frac{L}{c} \left[ \frac{\partial}{\partial E} \left( 1 - \frac{1}{\left( \frac{E}{m_n c^2} + 1 \right)^2} \right)^{-1} \right] \tag{C.12}
\]

Next we apply the chain rule for the contents of the parentheses in Eq. C.12, and come to Eq. C.13.

\[
\frac{\partial t}{\partial E} = \frac{L}{c} \left[ -1 \left( 1 - \frac{1}{\left( \frac{E}{m_n c^2} + 1 \right)^2} \right)^{-2} \left( -1 \frac{\partial}{\partial E} \left( \frac{E}{m_n c^2} + 1 \right)^{-2} \right) \right] \tag{C.13}
\]

Finally we come to the solution in Eq. C.14.

\[
\frac{\partial t}{\partial E} = \frac{L}{c} \left[ \left( 1 - \frac{1}{\left( \frac{E}{m_n c^2} + 1 \right)^2} \right)^{-2} \left( -2 \frac{E}{m_n c^2} \left( \frac{E}{m_n c^2} + 1 \right)^{-3} \right) \right] \tag{C.14}
\]

After simplification we come to Eq. C.15.

\[
\frac{\partial t}{\partial E} = -\frac{2L}{m_n c^3} \left[ \left( 1 - \frac{1}{\left( \frac{E}{m_n c^2} + 1 \right)^2} \right)^{-2} \left( \frac{E}{m_n c^2} + 1 \right)^{-3} \right] \tag{C.15}
\]
APPENDIX D
Cross Section Reconstruction with NJOY21

Following a resolved and unresolved resonance evaluation, the evaluated parameters must be stored in an efficient way: using the ENDF [28] format. The SAMMY/FITACS [51] program includes functions to output the fitted resonance parameters into “File 2” of this format, and NJOY21 [62] is designed to read this format and reconstruct cross section. The final product of the URR fit in this work was an ENDF format URR portion of File 2. The output contains energy dependent: average level spacing $D$, average neutron width $\langle \Gamma_n \rangle$, average radiation width $\langle \Gamma_\gamma \rangle$, and average inelastic width $\langle \Gamma_{n'} \rangle$ for every available total quantum angular momentum $J$ for the quantum angular moments of $L = 0, 1, 2$. This output was placed into the full ENDF file published in the JEFF-3.3 [16] evaluation and used as input to NJOY. NJOY processing typically involves recreating cross section for the URR in more than one module of the program. For the NJOY operation presented in this thesis, first the cross sections over all energies were reconstructed using the RECONR module, and then the PURR module is used to generate the cross section probability distributions in the URR region (2.5 to 100 keV). Using the output parameters from FITACS and processing with NJOY, the total cross section from RECONR has been plotted with a red line, and the infinite dilute cross section from PURR is plotted with black X’s in Fig. D.1. The cross section calculated by FITACS is plotted by green triangles.
It was clear that the cross section calculated by RECONR with the full evaluated file is non-physical, while the infinite dilute cross section calculated by PURR is more well behaved. Upon further investigation it was found that by removing a set of inelastic widths for a single spin state (L=1, J=2) the RECONR module calculated a much more reasonable cross section, shown by the blue line in Fig. D.1. This single set of inelastic widths were causing a problem because they were given degrees of freedom equal to zero (AMUX=0.0).

The JEFF-3.3 evaluators listed AMUX=0.0, and zeros for all the inelastic widths for that spin state. When placing the newly evaluated inelastic widths in the JEFF-3.3 file, the combination of the JEFF-3.3 value of AMUX=0.0 and the RPI values for the inelastic widths cause non-physical behavior in the URR. The NJOY21 output file did not (at the time of this writing) include any “warning” or “error” statements that commented on the incorrectly specified value in the URR. The NJOY developers have been informed of this behavior, and future versions of NJOY are expected to address the erroneous user input.
APPENDIX E
Covariance Matrix Input

Listed here in Tables E.1, E.2, and E.3 are the tabulated data necessary to reproduce the covariance for the validation transmission measurement. Table E.1 is similar to the input quantities required in the SAMMY IDC matrix file, along with most of the derivatives in Table E.3. All of the derivatives in Table E.3 are listed in descending energy which correspond to the energies listed in Table E.2. It should be noted that the uncertainty in transmission \( \Delta T \) is redundant, but listed here in Table E.2 for convenience.

Table E.1: The input covariance matrix \( C_{sy} \) for the systematic uncertainty variables found in the transmission equation (Eq. 4.1).

<table>
<thead>
<tr>
<th>( \Delta a )</th>
<th>( \Delta b )</th>
<th>( \Delta k_a )</th>
<th>( \Delta k_o )</th>
<th>( \Delta B_0_{Fp} )</th>
<th>( \Delta B_0_o )</th>
<th>( \Delta \alpha_1 )</th>
<th>( \Delta \alpha_2 )</th>
<th>( \Delta \alpha_3 )</th>
<th>( \Delta \alpha_4 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.38210e+03</td>
<td>1.72795e-01</td>
<td>0.0</td>
<td>0.0</td>
<td>5.77123e-04</td>
<td>6.62760e-03</td>
<td>0.0</td>
<td>0.0</td>
<td>2.56e-04</td>
<td>2.5e-05</td>
</tr>
<tr>
<td>1.72795e-01</td>
<td>2.65178e-05</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0e+00</td>
<td>1.0e+00</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>5.77123e-04</td>
<td>6.62760e-03</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0e+00</td>
<td>1.0e+00</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>2.56e-04</td>
<td>2.5e-05</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>1.0e+00</td>
<td>1.0e+00</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>6.40e-05</td>
<td>3.24e-04</td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>2.5e-05</td>
</tr>
</tbody>
</table>
Table E.2: The transmission, uncertainty on the transmission, and statistical uncertainty on the sample ($C_{Ta,i}$) and open ($C_{o,i}$) count rates.

<table>
<thead>
<tr>
<th>Energy [eV]</th>
<th>T</th>
<th>$\Delta T$</th>
<th>$\Delta C_{Ta,i}$</th>
<th>$\Delta C_{o,i}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.042935e+05</td>
<td>6.020775e-01</td>
<td>1.502753e-02</td>
<td>1.749919e+01</td>
<td>3.119723e+01</td>
</tr>
<tr>
<td>1.742499e+05</td>
<td>5.865481e-01</td>
<td>1.530542e-02</td>
<td>1.217420e+01</td>
<td>2.207235e+01</td>
</tr>
<tr>
<td>9.65381e+04</td>
<td>5.60185e-01</td>
<td>2.344247e-02</td>
<td>5.798835e+00</td>
<td>1.094345e+01</td>
</tr>
<tr>
<td>6.123407e+04</td>
<td>5.287610e-01</td>
<td>1.735712e-02</td>
<td>6.988538e+00</td>
<td>1.342885e+01</td>
</tr>
<tr>
<td>4.227036e+04</td>
<td>5.021612e-01</td>
<td>2.631982e-02</td>
<td>4.736515e+00</td>
<td>9.417466e+00</td>
</tr>
<tr>
<td>3.092397e+04</td>
<td>4.900893e-01</td>
<td>1.912396e-02</td>
<td>5.659308e+00</td>
<td>1.127112e+01</td>
</tr>
<tr>
<td>2.360094e+04</td>
<td>4.675339e-01</td>
<td>1.507341e-02</td>
<td>6.638204e+00</td>
<td>1.344494e+01</td>
</tr>
<tr>
<td>1.860165e+04</td>
<td>4.582755e-01</td>
<td>1.483866e-02</td>
<td>6.457516e+00</td>
<td>1.319365e+01</td>
</tr>
<tr>
<td>1.503763e+04</td>
<td>4.344542e-01</td>
<td>1.407572e-02</td>
<td>6.240187e+00</td>
<td>1.306350e+01</td>
</tr>
<tr>
<td>1.240777e+04</td>
<td>4.130323e-01</td>
<td>1.348575e-02</td>
<td>5.977681e+00</td>
<td>1.280843e+01</td>
</tr>
<tr>
<td>1.041198e+04</td>
<td>4.062710e-01</td>
<td>1.328117e-02</td>
<td>5.811017e+00</td>
<td>1.254492e+01</td>
</tr>
<tr>
<td>8.861682e+03</td>
<td>4.100818e-01</td>
<td>1.336032e-02</td>
<td>5.707302e+00</td>
<td>1.226485e+01</td>
</tr>
<tr>
<td>7.63514e+03</td>
<td>4.078846e-01</td>
<td>1.319744e-02</td>
<td>5.601712e+00</td>
<td>1.206601e+01</td>
</tr>
<tr>
<td>6.644041e+03</td>
<td>3.884243e-01</td>
<td>1.258477e-02</td>
<td>5.381499e+00</td>
<td>1.186000e+01</td>
</tr>
<tr>
<td>5.835190e+03</td>
<td>3.970554e-01</td>
<td>1.393857e-02</td>
<td>4.825200e+00</td>
<td>1.051985e+01</td>
</tr>
<tr>
<td>5.086131e+03</td>
<td>4.040754e-01</td>
<td>1.445548e-02</td>
<td>4.077495e+00</td>
<td>8.814296e+00</td>
</tr>
<tr>
<td>4.408543e+03</td>
<td>3.616516e-01</td>
<td>1.216291e-02</td>
<td>4.069118e+00</td>
<td>9.268613e+00</td>
</tr>
<tr>
<td>3.857863e+03</td>
<td>3.836367e-01</td>
<td>1.145471e-02</td>
<td>4.625882e+00</td>
<td>1.025292e+01</td>
</tr>
<tr>
<td>3.404270e+03</td>
<td>3.782777e-01</td>
<td>1.105713e-02</td>
<td>4.591101e+00</td>
<td>1.024888e+01</td>
</tr>
<tr>
<td>3.026214e+03</td>
<td>3.544523e-01</td>
<td>1.028832e-02</td>
<td>4.399201e+00</td>
<td>1.012905e+01</td>
</tr>
<tr>
<td>2.707808e+03</td>
<td>3.757980e-01</td>
<td>1.070352e-02</td>
<td>4.404474e+00</td>
<td>9.939092e+00</td>
</tr>
<tr>
<td>2.437132e+03</td>
<td>3.708814e-01</td>
<td>1.051355e-02</td>
<td>4.292388e+00</td>
<td>9.667872e+00</td>
</tr>
<tr>
<td>2.205103e+03</td>
<td>3.798402e-01</td>
<td>1.051259e-02</td>
<td>4.376198e+00</td>
<td>9.743090e+00</td>
</tr>
<tr>
<td>1.047764e+03</td>
<td>3.739412e-01</td>
<td>9.336665e-03</td>
<td>9.812306e-01</td>
<td>2.201038e+00</td>
</tr>
<tr>
<td>4.150293e+02</td>
<td>4.105222e-01</td>
<td>1.005748e-02</td>
<td>8.910313e-01</td>
<td>1.907201e+00</td>
</tr>
<tr>
<td>2.213887e+02</td>
<td>3.555513e-01</td>
<td>8.761434e-03</td>
<td>7.480531e-01</td>
<td>1.716702e+00</td>
</tr>
<tr>
<td>1.372256e+02</td>
<td>5.120513e-01</td>
<td>1.313945e-02</td>
<td>5.261448e-01</td>
<td>1.004973e+00</td>
</tr>
</tbody>
</table>
Table E.3: The first set of derivatives of the transmission equation given in Ch. 4 for the validation transmission. The derivatives occur at the energies listed in Table E.2.

<table>
<thead>
<tr>
<th>( \frac{\partial T_i}{\partial a} )</th>
<th>( \frac{\partial T_i}{\partial b} )</th>
<th>( \frac{\partial T_i}{\partial k_{ta}} )</th>
<th>( \frac{\partial T_i}{\partial k_{o}} )</th>
<th>( \frac{\partial T_i}{\partial \rho_{0}} )</th>
<th>( \frac{\partial T_i}{\partial B_0} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>-1.284273e-07</td>
<td>2.199647e-01</td>
<td>-2.072060e-02</td>
<td>-1.247540e-02</td>
<td>-5.187754e-05</td>
<td>-3.123430e-05</td>
</tr>
<tr>
<td>-2.589413e-07</td>
<td>5.112462e-01</td>
<td>-3.806998e-02</td>
<td>-2.23459e-02</td>
<td>-1.062738e-04</td>
<td>-6.233469e-05</td>
</tr>
<tr>
<td>-1.216769e-06</td>
<td>2.632637e+00</td>
<td>-1.638380e-01</td>
<td>-9.273535e-02</td>
<td>-5.101878e-04</td>
<td>-2.887757e-04</td>
</tr>
<tr>
<td>-7.058794e-07</td>
<td>1.618365e+00</td>
<td>-8.870881e-02</td>
<td>-4.690576e-02</td>
<td>-3.080713e-04</td>
<td>-1.628961e-04</td>
</tr>
<tr>
<td>-1.568602e-06</td>
<td>3.720693e+00</td>
<td>-1.820717e-01</td>
<td>-9.142932e-02</td>
<td>-7.051735e-04</td>
<td>-3.541108e-04</td>
</tr>
<tr>
<td>-9.792570e-07</td>
<td>2.360402e+00</td>
<td>-1.033291e-01</td>
<td>-5.064049e-02</td>
<td>-4.463191e-04</td>
<td>-2.187362e-04</td>
</tr>
<tr>
<td>-6.336500e-07</td>
<td>1.530651e+00</td>
<td>-6.154421e-02</td>
<td>-2.877400e-02</td>
<td>-2.964688e-04</td>
<td>-1.386092e-04</td>
</tr>
<tr>
<td>-6.469331e-07</td>
<td>1.548695e+00</td>
<td>-5.696035e-02</td>
<td>-2.610353e-02</td>
<td>-3.060086e-04</td>
<td>-1.402362e-04</td>
</tr>
<tr>
<td>-6.366945e-07</td>
<td>1.496827e+00</td>
<td>-5.173138e-02</td>
<td>-2.247491e-02</td>
<td>-3.099446e-04</td>
<td>-1.346567e-04</td>
</tr>
<tr>
<td>-6.417668e-07</td>
<td>1.470470e+00</td>
<td>-4.801397e-02</td>
<td>-1.983132e-02</td>
<td>-3.208240e-04</td>
<td>-1.325107e-04</td>
</tr>
<tr>
<td>-6.604019e-07</td>
<td>1.465338e+00</td>
<td>-4.468229e-02</td>
<td>-1.815312e-02</td>
<td>-3.329689e-04</td>
<td>-1.352756e-04</td>
</tr>
<tr>
<td>-6.915727e-07</td>
<td>1.477846e+00</td>
<td>-4.175450e-02</td>
<td>-1.712276e-02</td>
<td>-3.470089e-04</td>
<td>-1.423020e-04</td>
</tr>
<tr>
<td>-7.094783e-07</td>
<td>1.453189e+00</td>
<td>-3.851596e-02</td>
<td>-1.571007e-02</td>
<td>-3.569827e-04</td>
<td>-1.456078e-04</td>
</tr>
<tr>
<td>-7.134779e-07</td>
<td>1.394903e+00</td>
<td>-3.560709e-02</td>
<td>-1.383066e-02</td>
<td>-3.680545e-04</td>
<td>-1.429613e-04</td>
</tr>
<tr>
<td>-9.249932e-07</td>
<td>1.719827e+00</td>
<td>-4.093470e-02</td>
<td>-1.625334e-02</td>
<td>-4.718852e-04</td>
<td>-1.873646e-04</td>
</tr>
<tr>
<td>-1.053275e-06</td>
<td>1.842881e+00</td>
<td>-4.083238e-02</td>
<td>-1.649936e-02</td>
<td>-5.325342e-04</td>
<td>-2.151840e-04</td>
</tr>
<tr>
<td>-8.892583e-07</td>
<td>1.447599e+00</td>
<td>-3.174938e-02</td>
<td>-1.148221e-02</td>
<td>-4.752314e-04</td>
<td>-1.718682e-04</td>
</tr>
<tr>
<td>-7.374006e-07</td>
<td>1.112287e+00</td>
<td>-2.228142e-02</td>
<td>-8.547970e-03</td>
<td>-3.827716e-04</td>
<td>-1.468452e-04</td>
</tr>
<tr>
<td>-7.303505e-07</td>
<td>1.017178e+00</td>
<td>-1.936383e-02</td>
<td>-7.329400e-03</td>
<td>-3.817818e-04</td>
<td>-1.441959e-04</td>
</tr>
<tr>
<td>-8.067580e-07</td>
<td>8.688340e-01</td>
<td>-1.428774e-02</td>
<td>-5.299058e-03</td>
<td>-4.258618e-04</td>
<td>-1.579442e-04</td>
</tr>
<tr>
<td>-8.011913e-07</td>
<td>7.880268e-01</td>
<td>-1.221792e-02</td>
<td>-4.640856e-03</td>
<td>-4.179548e-04</td>
<td>-1.587560e-04</td>
</tr>
<tr>
<td>-5.090843e-06</td>
<td>3.929321e-03</td>
<td>-1.353996e-05</td>
<td>-6.933154e-06</td>
<td>-2.263320e-03</td>
<td>-1.158936e-03</td>
</tr>
</tbody>
</table>
Table E.4: The second set of derivatives of the transmission equation given in Ch. 4 for the validation transmission. The derivatives occur at the energies listed in Table E.2.

<table>
<thead>
<tr>
<th>$\frac{\partial T_i}{\partial x_1}$</th>
<th>$\frac{\partial T_i}{\partial x_2}$</th>
<th>$\frac{\partial T_i}{\partial x_3}$</th>
<th>$\frac{\partial T_i}{\partial x_4}$</th>
<th>$\frac{\partial T_i}{\partial C_{T_{u_1}}}$</th>
<th>$\frac{\partial T_i}{\partial C_{C_{u_1}}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.142619e-01</td>
<td>-1.166570e-02</td>
<td>6.208473e-01</td>
<td>-1.835132e-02</td>
<td>5.187754e-05</td>
<td>3.123430e-05</td>
</tr>
<tr>
<td>5.817848e-01</td>
<td>-4.994306e-02</td>
<td>5.999422e-01</td>
<td>-6.899837e-02</td>
<td>3.080713e-04</td>
<td>1.628961e-04</td>
</tr>
<tr>
<td>5.527268e-01</td>
<td>-5.817429e-02</td>
<td>5.675125e-01</td>
<td>-7.449217e-02</td>
<td>4.463191e-04</td>
<td>2.187362e-04</td>
</tr>
<tr>
<td>5.051480e-01</td>
<td>-3.464939e-02</td>
<td>5.117178e-01</td>
<td>-4.232656e-02</td>
<td>2.964688e-04</td>
<td>1.386092e-04</td>
</tr>
<tr>
<td>3.842788e-01</td>
<td>-1.787490e-02</td>
<td>3.808450e-01</td>
<td>-1.689034e-02</td>
<td>4.752314e-04</td>
<td>1.718682e-04</td>
</tr>
<tr>
<td>4.000089e-01</td>
<td>-1.254444e-02</td>
<td>3.981785e-01</td>
<td>-1.257406e-02</td>
<td>3.827716e-04</td>
<td>1.468452e-04</td>
</tr>
<tr>
<td>3.929973e-01</td>
<td>-1.090184e-02</td>
<td>3.909878e-01</td>
<td>-1.077493e-02</td>
<td>3.817818e-04</td>
<td>1.444195e-04</td>
</tr>
<tr>
<td>5.346921e-01</td>
<td>-7.622997e-06</td>
<td>5.275913e-01</td>
<td>-1.019867e-05</td>
<td>2.263320e-03</td>
<td>1.158936e-03</td>
</tr>
</tbody>
</table>
APPENDIX F
Determining Uncertainty on Monitor Counts

The neutron production intensity at the RPI Linac is monitored by fission chambers placed approximately 9 meters away from the neutron production target, in the flight path tunnel. As TOF counts are needed for multiple samples (open, sample in, reference sample, etc...) the samples are cycled, meaning each sample is given \( \approx 15 \) minutes of beam time before the next sample is put in the beam. This ensures that, if the beam intensity changes over the run, each sample will be exposed to approximately the same beam intensity. The intensity variation is also corrected by the monitor count rates. The counts as measured by the monitors are recorded per cycle and are typically on the order of \( 10^5 \) or greater, making the uncertainty in count rate nearly negligible (\( \sqrt{C}/C < 0.3\% \)). Another measure of the uncertainty in the monitor rates, however, is the deviation of the monitor rates away from the total detector (with which we are measuring TOF) counts. As shown in Eq. F.1, the standard deviation of the normalized monitor-detector ratios \( x \) have been defined as \( \sigma_{mon} \).

\[
\sigma_{mon} = \sqrt{\frac{1}{N-1} \sum (x - \langle x \rangle)^2}, \quad x \equiv \frac{C_{mon}/C_{det}}{\langle C_{mon}/C_{det} \rangle} \quad \text{(F.1)}
\]

The fractional uncertainty in the standard deviation of \( x \) is taken as the uncertainty of that monitor. In Fig. G.1, \( x \) has been plotted as a function of cycle for the \(^{181}\)Ta and Open from the thick-sample transmission measurement.
Figure F.1: The values $\frac{C_{\text{mon}}}{C_{\text{det}}}$ have been plotted as a function of cycle number for $^{181}$Ta and Open from the thick-sample transmission measurement. The dashed horizontal lines indicate the 2-sigma uncertainty for Ta (blue) and Open (red).
APPENDIX G
Copyright Permissions

Permission was obtained from Dr. Nicholas Thompson in order to use a figure from his thesis. This figure was important to establish the previous work done to measure $^{181}$Ta before this publication (Brown), as well as demonstrate the importance of the RRR.

Figure G.1: The written permission from Dr. Nicholas Thompson for use of his copyrighted figure.