

Evaluation of Tagging Techniques for Gamma-decay Probabilities Using the Surrogate Method

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To my loving parents, for which all of this is possible

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Few are those who see with their own eyes and feel with their own hearts.

- Albert Einstein

Abstract

A detailed analysis of the statistical and discrete γ [gamma]-decay tagging techniques was conducted using the absolute surrogate and surrogate ratio method (SRM) to obtain the $^{92}\text{Mo}(n,\gamma \text{ [gamma]})$ cross section in an equivalent neutron energy range of 80 to 880 keV. Excited ^{93}Mo and ^{95}Mo nuclei were populated using (d,p) reactions on ^{92}Mo and ^{94}Mo targets, respectively. The absolute surrogate $^{92}\text{Mo}(n,\gamma \text{ [gamma]})$ cross sections disagreed with evaluated neutron capture cross section data by as much as a factor of 4 using the statistical tagging approach, whereas the discrete γ [gamma]-decay tag absolute surrogate cross section disagreed with the evaluated neutron capture cross section by as much as a factor of 2, with both, statistical and discrete γ [gamma]-decay tagging techniques showing an unfavorable trend with the results. The surrogate cross sections obtained via the SRM, in comparison to the absolute surrogate method, offered a more favorable trend with the evaluated data as well as a more agreeable measurement with the evaluated ^{92}Mo cross section. The experimental results suggest that the discrete and statistical γ [gamma]-decay channel tagging approaches may sample different contributions of the γ [gamma]-cascade from the residual nucleus for the near spherical nuclei probed in this experiment. This work serves as the first evaluation of the surrogate method in the determination of neutron capture cross sections on spherical and quasi-spherical nuclei in the mass-90 region and provides a possible pathway to extend the SRM to a broader mass range.

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Chapter 1

Introduction

The surrogate method is an experimental technique for the determination of neutron-induced reaction cross sections through the use of light-ion direct reactions, termed “surrogate” reactions (e.g., (d, p) , $({}^3\text{He}, {}^4\text{He})$, (p, p') , etc.), originally employed in the early 1970s [1, 2]. The surrogate method has had a recent resurgence in the determination of neutron-induced fission cross sections for both stable and radioactive nuclei with an associated uncertainty on the order of 10% or less in some cases [3]. The surrogate method boasts the advantage of being able to use a stable target to access the same compound nucleus as a neutron-induced reaction on a highly radioactive target, which avoids some of the difficulties presented when dealing with the measurement of rapidly decaying nuclei.

The absolute surrogate method (ASM) and the surrogate ratio method (SRM) have been employed in the literature in order to determine the surrogate cross section measurement. Boyer *et al.* employed the ASM via the ${}^{232}\text{Th}({}^3\text{He}, p){}^{234}\text{Pa}^*$ transfer reaction in order to extract ${}^{233}\text{Pa}(n, \gamma)$ cross sections in the neutron energy range of 0 to 1 MeV [4]. The SRM is an expansion of the ASM that employs a ratio of two similar target nuclei in order to measure the desired reaction cross section initially introduced by Plettner *et al.* [5]. Burke *et al.* expanded on this work by using the SRM to extract a cross section for ${}^{237}\text{U}(n, f)$ reaction over an equivalent neutron

energy range of 0 to 20 MeV [6]. The SRM eliminates the need to accurately measure the total number of direct reaction “surrogate” events, which has been the largest source of uncertainty in recent surrogate experiments by using a ratio of an unknown cross section to a known one. Both the ASM and SRM are explained in further detail in Sec. 2. The surrogate method was recently expanded to determine neutron capture cross sections in the rare-earth region as well as spherical and near-spherical nuclei [7, 8].

In the realm of nuclear engineering, radiative neutron-capture cross sections are extremely important in many applications within the nuclear power industry. Reductions of uncertainties in nuclear data has the potential to generate a considerable economic impact on both the front end (mining, conversion, enrichment and fuel fabrication) as well as the back end (reprocessing and waste management) of the fuel cycle, saving the nuclear industry millions of dollars [9]. In addition to nuclear industry applications, radiative neutron-capture reactions also play an important role in many areas of basic and applied nuclear science. For example, in order to uncover properties of the astrophysical environments relevant to the synthesis of heavy elements by astrophysical s-process, more accurate (n,γ) cross sections are required [10]. The surrogate method is an experimental technique that can be applied in cases where conventional neutron-induced measurements are difficult, which serves as a valuable method for increasing the experimental cross section database.

As illustrated in Fig. 1.1, the desired reaction investigated in this experiment is the $^{92}\text{Mo}(n,\gamma)$ reaction which proceeds through the formation of the ^{93}Mo compound nucleus. In this case a surrogate (d,p) reaction was employed on a ^{92}Mo target to populate the ^{93}Mo residual nucleus which is assumed to damp into a ^{93}Mo compound nucleus. The P_n term in Fig. 1.1 represents the probability of neutron emission from the compound nucleus, which becomes relevant above the neutron separation energy. The P_γ term represents the probability of γ -decay from the compound nucleus. In order to obtain a surrogate cross section using the $^{92}\text{Mo}(d,p\gamma)$ reaction, the probability of γ -decay from the compound nucleus formed in the surrogate reaction

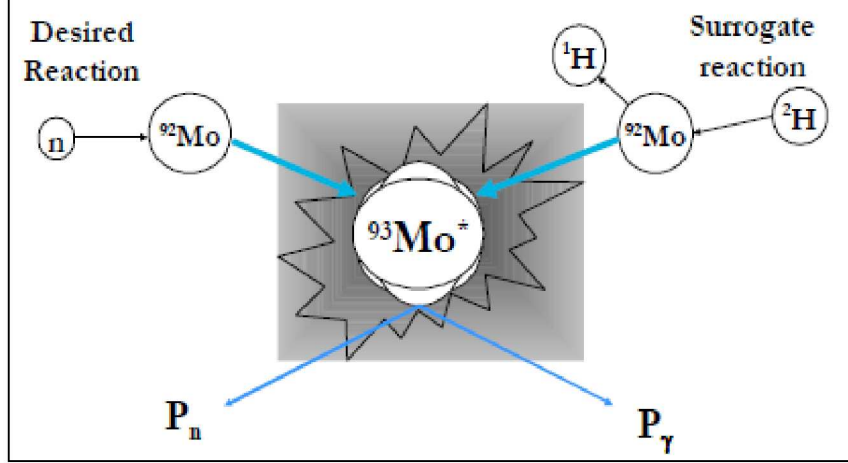


Figure 1.1: Schematic of the surrogate technique

must be extracted. This term is represented by $P_{(d,p\gamma)}(E_x)$ and represents the γ -decay probability for the compound nucleus formed via the surrogate reaction as a function of excitation energy, E_x , and is what is subsequently measured in order to determine the surrogate neutron capture cross section. How this term is used to determine the cross section will be discussed in detail in Sec. 2.

The application of the surrogate method described in the text serves as an extension of the surrogate method to spherical and near-spherical nuclei. However, the main focus of this analysis is the direct comparison of two different approaches used to tag the γ -decay channel (i.e., statistical and discrete tagging methods) in the indirect determination of neutron capture cross sections using the surrogate method. The successful tagging of the γ -decay channel leads to the quantification of the surrogate γ -decay probability, $P_{(d,p\gamma)}(E_x)$, and allows for the determination of the surrogate neutron capture cross section.

The discrete tagging approach involves using a known transition of the compound nucleus to identify the γ -decay channel. The intensity of that transition as a function of energy is used to quantify the surrogate γ -decay probability. The validity of this technique relies on the inclusion of the known transition in the γ -cascade of the compound nucleus. This method yields relatively low statistics. However, the use of a

known transition in the compound nucleus provides a unique signature for the isotope of interest which accurately tags the compound nucleus and avoids any potential contributions from impurities in the target.

There are many variations of the discrete γ -decay channel tagging approach. A model dependent version of the discrete tagging approach was used by Hatarik, *et al.* by using the $^{171,173}\text{Yb}(d,p)$ reactions to excite $^{172,174}\text{Yb}$ residual nuclei in order to obtain the ^{171}Yb to ^{173}Yb neutron capture cross section ratio [11]. The known discrete transition used to determine γ -decay probability was the $4^+ \rightarrow 2^+$ transition with contributions from the $6^+ \rightarrow 4^+$ transition removed through the use of a Monte Carlo simulation. This method yielded surrogate ratio cross sections that disagreed with the directly-measured data by as much as 30%, with a shape that did not trend well with the directly-measured data. Another discrete tagging approach was developed by Allmond, *et al.* to obtain the $^{235}\text{U}(n,\gamma)$ cross section relative to the $^{235}\text{U}(n,f)$ cross section using the $^{235}\text{U}(d,p\gamma)$ to $^{235}\text{U}(d,pf)$ probability ratio [12]. This variation of the discrete tagging approach utilized empirical data on the low-lying level scheme of ^{236}U to obtain the γ -channel yield from multiple γ -cascade paths. The surrogate ratio cross section data had a shape that trended well with the evaluated cross section data with an average deviation of 23%. However, a drawback of this method is that the heavy reliance of knowledge of the low-lying level scheme of the relevant compound nucleus limits the applicability of this variation of the discrete γ -decay channel tagging technique to only well-studied nuclei.

In the statistical tagging approach the γ -decay probability is obtained by determining the total number of γ -rays in a given energy range gate as a function of excitation energy. The statistical approach is based on the assumption that the γ -rays within the determined energy range are primary transitions in the compound nucleus. This approach excels in the fact that low-resolution, high-efficiency γ -ray detectors may be employed which results in significantly higher statistics. However, a hindrance of the statistical technique is the introduction of additional uncertainty arising from potential target contamination. A recent application of the statistical γ -decay channel

tagging approach employed by Goldblum, *et al.* obtained the $^{161}\text{Dy}(n,\gamma)$ cross section through the use of a low-resolution NaI multi-detector array [13]. A γ -ray energy threshold was applied to contain primary transitions from higher excitation energies in order to experimentally determine the surrogate $^{161}\text{Dy}(n,\gamma)$ cross section, which showed general agreement with the directly-measured $^{161}\text{Dy}(n,\gamma)$ cross section data over the entire equivalent neutron energy range. However, a large uncertainty was introduced in the data due to target contamination as the target was only 95% pure.

The application of the surrogate technique on the spherical and near-spherical nuclei considered in this text allows for a direct comparison of the discrete γ -decay channel tagging technique to the statistical γ -decay channel technique using both the SRM and ASM. A recent work has been published which compares both the statistical and discrete γ -decay channel tagging techniques for $^{157,155}\text{Gd}$ nuclei which concluded that both the discrete and statistical γ -decay channel tagging technique showed similar results, with both techniques suffering from the angular-momentum effects observed in other surrogate reaction experiments [14, 15]. This thesis coincides with work recently published on the application of the surrogate method to spherical and near-spherical nuclei and serves as review of the methods used for the measurement [8].

Chapter 2

Theory

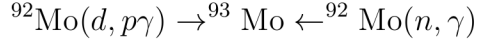
There are two standard techniques commonly used within the literature in the evaluation of cross sections using the surrogate technique, the absolute surrogate surrogate (ASM) and the surrogate ratio method (SRM). However, both techniques discussed in the majority of applications of the surrogate method rely on the validity of the Weisskopf-Ewing limit of the Hauser Feshbach theory [16].

2.1 Absolute Surrogate Method

Within the Weisskopf-Ewing limit, decay probabilities can be considered independent of total angular momentum and parity of the states populated in the compound nucleus via the surrogate reaction. This assumption reduced the Hauser-Feshbach formula to an expression for the excitation-energy dependent neutron-induced γ -decay cross section in terms of the surrogate reaction γ -decay probability. Thus, within the Weisskopf-Ewing limit, the absolute surrogate cross section, relevant for this work can be expressed by the following:

$$\sigma_{(n,\gamma)}(E_n) = \sigma_n^{CN}(E_n)P_{(d,p\gamma)}(E_x), \quad (2.1)$$

where E_n is the incident neutron energy, E_x is the excitation energy in the compound nucleus, $\sigma_n^{CN}(E_n)$ is the neutron-induced compound nuclear formation cross section, and $P_{(d,p\gamma)}(E_x)$ is the γ -decay probability for the compound nucleus formed via the surrogate (d, p) reaction. The compound-nuclear formation cross section, $\sigma_n^{CN}(E_n)$, must be calculated using an optical model. The Koning-Delaroche global optical potential served as the model by which $\sigma_n^{CN}(E_n)$ was calculated [17]. This calculation was provided by theorists at Lawrence Livermore National Laboratory (LLNL), with an approximated uncertainty of 10% over the entire equivalent neutron energy range examined [18]. The surrogate reaction used for the cross section measurement via the ASM is displayed below, showing the surrogate reaction populating the same residual nucleus as that of the compound nucleus populated by the desired reaction.



In Eq. 2.1 the Weisskopf-Ewing approximation allows for an expression for the desired reaction cross section to be given in terms of the surrogate reaction decay probability, $P_{d,p\gamma}(E_x)$ expressed by the following:

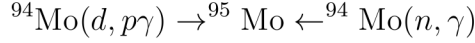
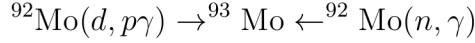
$$P_{(d,p\gamma)}(E_x) = \frac{N_{(d,p\gamma)}(E_x)}{N_{(d,p)}(E_x)} \quad (2.2)$$

where $N_{(d,p\gamma)}(E_x)$ is the number of γ -decay events in coincidence with the surrogate reaction ejectile and $N_{(d,p)}(E_x)$ is the total number of surrogate reaction events, both as functions of excitation energy, E_x . $N_{(d,p)}(E_x)$ is directly measured from the experimental data. In order to quantify the $N_{(d,p\gamma)}(E_x)$ term a 2-D proton-gated matrix of compound nuclear excitation energy and γ -ray energy was generated. The 2-D matrix was used to create 100-keV bins in the range of 7 – 9 MeV of excitation energy in the compound nucleus. The γ -ray spectra from the de-excitation of the compound nucleus within this energy range were extracted for each 100-keV bin. These γ -ray spectra were used for the measurement of $N_{(d,p\gamma)}(E_x)$ using the two γ -decay channel tagging techniques discussed in 2.3.

2.2 Surrogate Ratio Method

In contrast to the ASM, the SRM measures the decay of two compound nuclei relative to each other and employs the use of evaluated data of the cross section for the fiducial nucleus in order to determine a cross section for the nucleus of interest. This allows for correlated errors in the measurement to cancel in the ratio. Possible correlated errors can be incurred through experimental errors (e.g, background radiation and uncertainties in the detection efficiency) and can cancel in the ratio if the experimental set up for each target nucleus is similar. Most notably, simulations have shown that deviations in the angular momentum population for the two compound nuclei employed in the SRM may cancel if the two nuclei are similar, showing a less pronounced sensitivity in the angular momentum population of the compound nuclei in the SRM than the ASM [19].

A deuteron beam was employed on both the ^{92}Mo and ^{94}Mo target to induce a $(d, p\gamma)$ reaction that populated the same residual nuclei as the compound nucleus populated by the desired (n, γ) illustrated by the following reactions:



The cross section obtained using these reactions for the SRM can be expressed by the following:

$$\sigma_{(n, \gamma)}^{(^{92}\text{Mo})}(E_n) = \sigma_{(n, \gamma)}^{(^{94}\text{Mo})}(E_n) \frac{\sigma_n^{(^{93}\text{Mo})}(E_n) P_{(d, p\gamma)}^{(^{92}\text{Mo})}(E_x)}{\sigma_n^{(^{95}\text{Mo})}(E_n) P_{(d, p\gamma)}^{(^{94}\text{Mo})}(E_x)} \quad (2.3)$$

The measured $\sigma_{(n, \gamma)}^{(^{92}\text{Mo})}(E_n)$ cross section is then essentially a ratio of two absolute surrogate measurements of the respective nuclei multiplied by the evaluated ENDF/B-VII.0 $^{94}\text{Mo}(n, \gamma)$ cross section [20]. In Eq. 2.3, the $\sigma_n^{(^{93, 95}\text{Mo})}(E_n)$ terms represent the calculated compound nuclear formation cross sections for the respective residual nuclei

as a function of incident neutron energy. The $P_{(d,p\gamma)}^{(92,94\text{Mo})}(E_x)$ terms represent the γ -decay probabilities of the two residual nuclei in coincidence with a surrogate reaction, which are each measured through the two different γ -decay tagging techniques discussed in the following section.

2.3 γ -decay Tagging Technique

In order to complete the measurement of the γ -decay probability for the compound nucleus formed via the surrogate reaction, $P_{d,p\gamma}(E_x)$, two different approaches were employed to determine the number of γ -decay events in coincidence with the surrogate reaction ejectile: $N_{d,p\gamma}(E_x)$.

2.3.1 Discrete γ -decay Tagging Technique

The discrete γ -decay tagging approach employed in this work involves integrating the peak area of the lowest-lying transition in the ^{93}Mo residual nucleus at 943.28 keV ($1/2^+ \rightarrow 5/2^+$), which tags the γ -decay channel. The lowest lying transition is used in an attempt to determine a transition that is assumed to collect all parallel decay paths from the compound nucleus. For the SRM, the lowest-lying transition in the ^{95}Mo residual nucleus at 204.1 keV ($3/2^+ \rightarrow 5/2^+$) was used to tag the γ -decay channel. The formal expression of the discrete γ -ray tagging approach is as follows:

$$N_{(d,p\gamma)}^{disc}(E_x) = \int_{peak} \frac{N'_{(d,p\gamma)}(E_x, E_\gamma)}{\epsilon(E_\gamma)} dE_\gamma \quad (2.4)$$

where $N'_{d,p\gamma}(E_x, E_\gamma)$ represents the number of proton- γ coincident events measured from γ -ray spectra from 100 keV bins of excitation energy extracted from the 2-D proton- γ coincident matrix, discussed in detail in Sec. 3. $N'_{d,p\gamma}(E_x, E_\gamma)$ is then divided by the efficiency of the detector array, $\epsilon(E_\gamma)$ and integrated for the peak area of the lowest-lying transition of the compound nucleus above background. Figure 2.1 shows an example γ -ray spectra obtained from the 2-D proton- γ coincident matrix with the

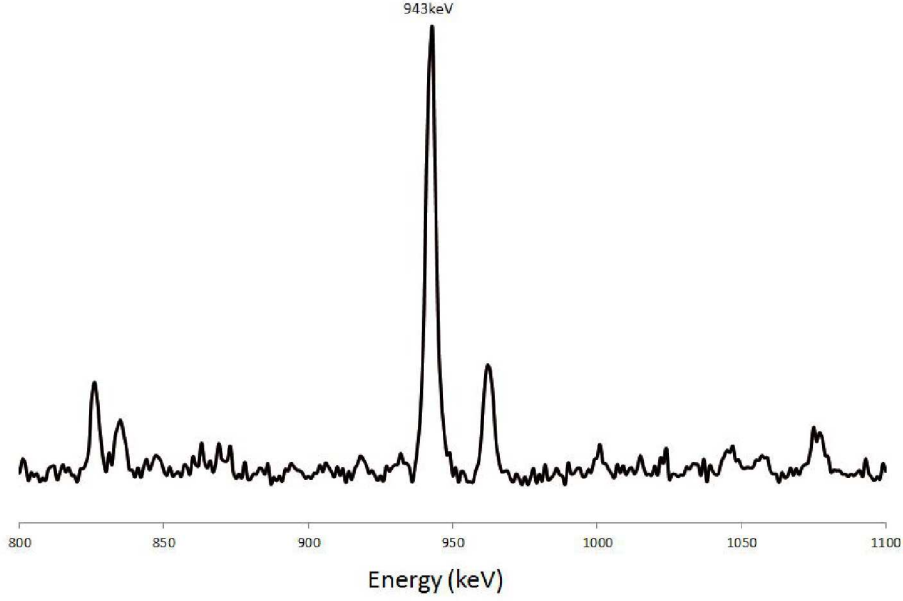


Figure 2.1: An example of a γ -ray spectrum extracted from a 100 keV bin between 8000-8100 keV in excitation energy in the ^{93}Mo compound nucleus with the low-lying transition of interest noted. The peak area for this transition was calculated using a peak fitting routine in gf3.

low-lying transition in ^{93}Mo at 943 keV labeled, which was integrated for the peak area in order to determine $N_{(d,p\gamma)}^{disc}(E_x)$.

2.3.2 Statistical γ -decay tagging technique

The assumption behind the statistical tagging technique lies within determining an energy range in an attempt to account for only the primary transitions emitted from the de-excitation γ -cascade from the compound nucleus in its decay to the ground state. For example, in the energy range of 8 – 9 MeV the ^{93}Mo compound nucleus can emit on average 3 γ -rays on its de-excitation down to the ground state [21]. The primary transition is the first, higher energy γ -ray emitted from the compound nucleus. In order to capture only one γ -ray for each $(d,p\gamma)$ decay cascade (a requirement in order to perfectly describe the γ -decay channel), the statistical tagging technique incorporated a calculated γ -ray energy range in an attempt to isolate only

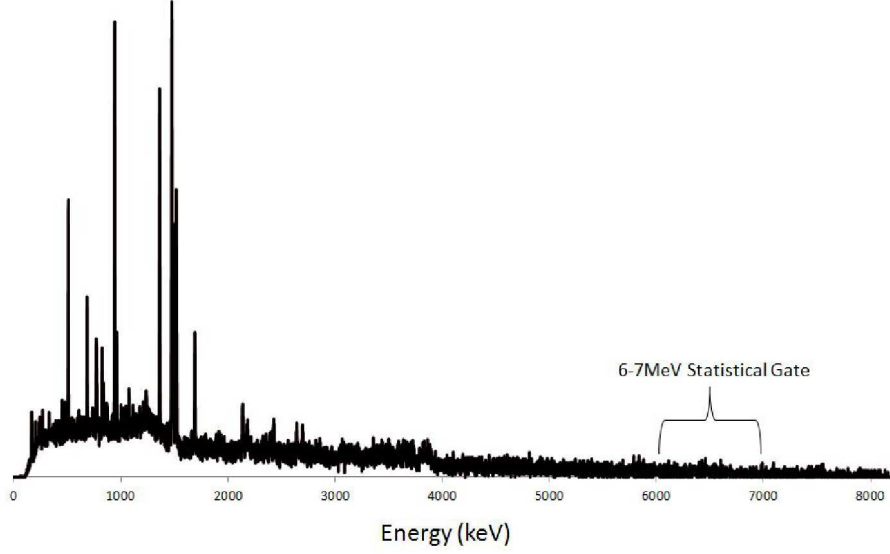


Figure 2.2: An example of a γ -ray spectrum extracted from a 100 keV bin between 8000-8100 keV in excitation energy in the ^{93}Mo compound nucleus. The bracket from 6000 keV to 7000 keV represents the statistical gate.

and all the primary transitions emitted from each γ -decay cascade from the populated surrogate compound nucleus.

Through a statistical model from Dossing and Vigezzi, the 6 – 7 MeV range was determined as an appropriate range to account for solely the primary transition emitted from the compound nucleus [22]. A formal expression for the statistical tagging technique is as follows:

$$N_{(d,p\gamma)}^{stat}(E_x) = \int_{6 \text{ MeV}}^{7 \text{ MeV}} \frac{N'_{(d,p\gamma)}(E_x, E_\gamma)}{\epsilon(E_\gamma)} dE_\gamma \quad (2.5)$$

where $N'_{(d,p\gamma)}(E_x, E_\gamma)$ represents the number of (d,p γ) events measured from 2-D particle- γ coincident matrix as a function of excitation energy in the compound nucleus and γ -ray energy. This term is then divided by the efficiency of the detector as a function of γ -ray energy, $\epsilon(E_\gamma)$, and integrated over the 6-7 MeV region, after background subtraction, to determine $N_{(d,p\gamma)}^{stat}(E_x)$. Contributions within the 6-7 MeV range from Compton-scattering are subtracted using a shelf model. Figure. 2.2 illustrates a γ -ray spectrum extracted from the 2-D proton- γ coincident matrix from a

100 keV energy bin between 8000-8100 keV in excitation energy in the ^{93}Mo compound nucleus, the bracket serves as a visual representation of the 6 – 7 MeV range used for the statistical γ decay tagging technique.

Chapter 3

Materials and Methods

3.1 Equipment and Setup

The experiment was conducted at the 88-inch Cyclotron at Lawrence Berkeley National Laboratory using an 11 MeV deuteron beam which subsequently struck one of two metallic foil targets: a ^{92}Mo target for the ASM, and a ^{94}Mo target (which was used in conjunction with the ^{92}Mo target for the SRM). Using the target wheel displayed in Fig 3.1, targets were changed without breaking the vacuum seal containing the Silicon Telescope Array for Reaction Studies discussed in the following section.

3.1.1 Silicon Telescope Array for Reaction Studies

After successful collisions of the deuteron beam on the target foil, charged particle ejectiles were detected using the Silicon Telescope Array for Reaction Studies (STARS), illustrated schematically in Fig. 3.2. The STARS array consists of a particle telescope with two double-sided Micron Semiconductor S2-type silicon detectors, a $150\text{ }\mu\text{m}$ ΔE (front) detector and a $1000\text{ }\mu\text{m}$ E (back) detector. The detectors are set up in a telescope array in order to perform particle identification. The telescope array allows for the ejectile to pass through the $150\text{ }\mu\text{m}$ ΔE detector and deposit only a



Figure 3.1: Target wheel

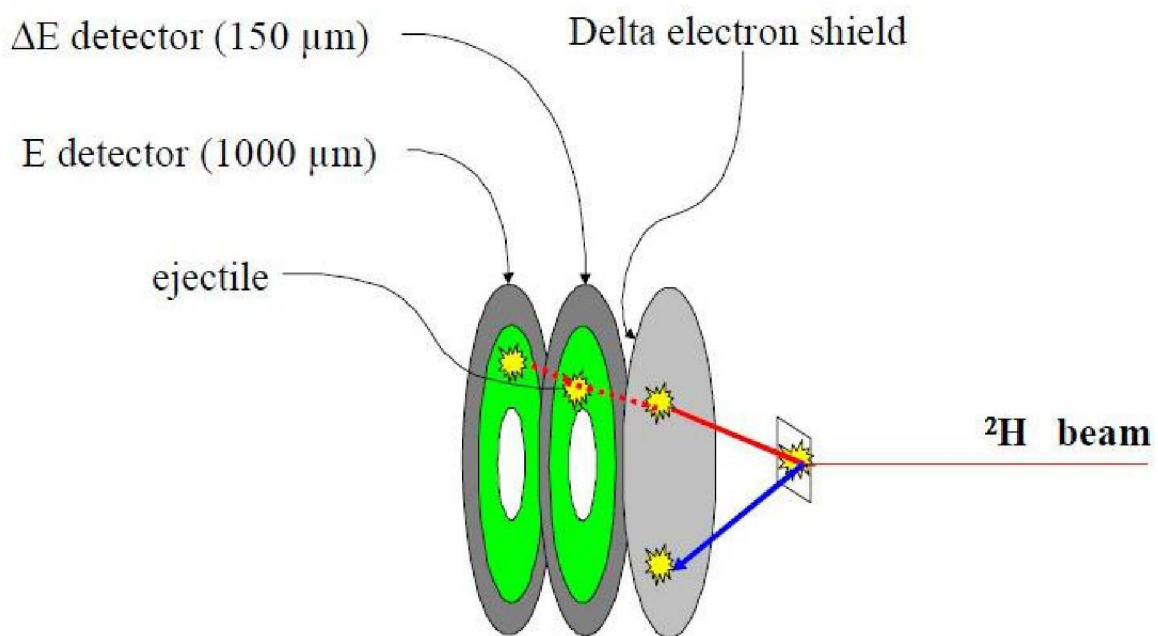


Figure 3.2: Particle telescope array

fraction of its energy, the ejectile then passes through the 1000 μm E detector, which has a thickness necessary to stop the ejectile. This yields the information necessary to identify the particle of interest. The silicon detectors were segmented into the electrical equivalent of 24 annular rings on one side and 8 sectors on the other side in order to determine the polar angle of detection of the ejectile (further discussed in Sec. 3.2.1).

3.1.2 Livermore Berkeley Array for Collaborative Experiments

The γ -ray spectra were detected using the Livermore BERkeley Array for Collaborative Experiments (LIBERACE). Figure 3.3 shows the STARS telescope array housed in a vacuum-sealed aluminum cannister shown in the middle of the picture along the beam line, surrounded by five high-purity germanium (HPGe) detectors. The HPGe detectors were surrounded by bismuth germanate oxide (BGO) scintillator shields, which allowed for Compton suppression. Compton suppression was achieved by removing Compton scattered events that generate an initial signal in the HPGe detector and then coincidentally generate a signal in the BGO shield as they scatter out of the detector, within a pre-set coincident time window.

Figure 3.4 shows the clover array for one of the HPGe detectors, which differs from a single crystal germanium detector in that it consists of four crystals mounted in a common cryostat [23]. This allows for a greater energy absorption efficiency, in that, the full energy peak of a Compton scattered photon absorbed in adjacent crystals can be determined. The full energy peak can be obtained by summing the energies deposited in the crystals fired from a Compton scattering event followed by a photoelectric event detected in adjacent clover crystals within the given HPGe detector. This full energy peak is summed from the relevant crystals fired during the Compton scattering of a single photon by utilizing a “gamma addback” routine in the data analysis code using timing considerations.

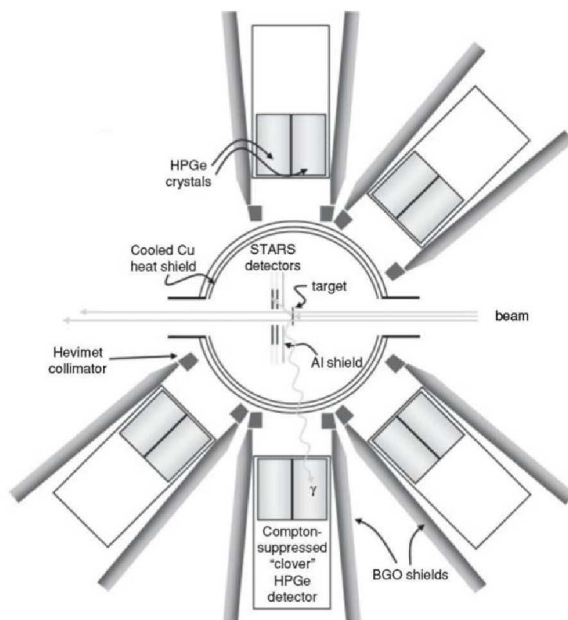


Figure 3.3: Experimental setup with HPGe Clover detectors surrounding the STARS particle telescope along the beam line.

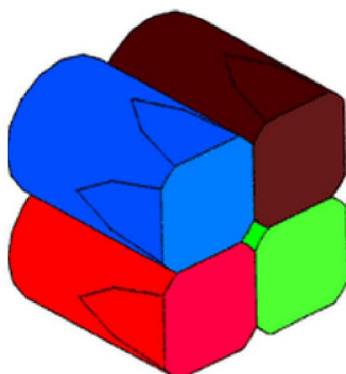


Figure 3.4: Four germanium crystals “leaves” in HPGe clover array

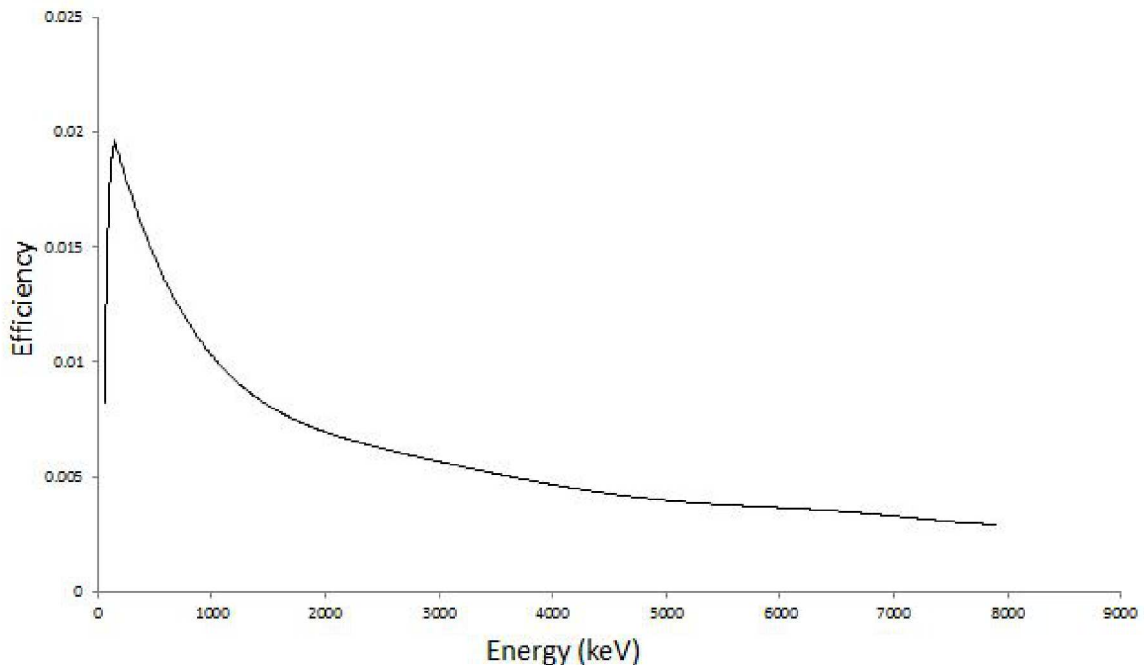


Figure 3.5: HPGe detector efficiency curve as a function of γ -ray energy.

The HPGe detector efficiency calibration was performed using a standard ^{152}Eu γ -ray source as well as particle- γ and particle- γ - γ coincidences using the ^{92}Mo experimental data [24]. The medium and high-energy γ -ray efficiency was determined using a carbon target to generate the $^{12}\text{C}(d, p\gamma)^{13}\text{C}$ and $^{13}\text{C}(d, p\gamma)^{14}\text{C}$ reactions in order to measure the medium and high energy transitions created in these reactions. This ultimately yielded an efficiency curve for the HPGe detector array, Fig. 3.5, as a function of γ -ray energy which was fit using polynomial expressions and incorporated into the measurement data.

3.2 Data analysis

The Sparrow Corporation Kmax data acquisition system generated raw binary data files which were parsed and converted from binary to decimal format using TSCAN v2.6 with a custom analysis code, in order to generate histogram files. The histogram files were capable of being arranged into one- or two-dimensional data arrays [25].

The data array matrices from the histogram file were written as spectra or text files using RGMT, a supporting program of TSCAN v2.6. The one- and two- dimensional data arrays were visualized through the use of the Display, Analysis and Manipulation Module (DAMM) [26]. Least-squares peak-fitting for detector calibration and peak-area quantification was accomplished using the Radware program gf3 [27].

3.2.1 Particle Identification

Particle identification of the direct reaction ejectiles (i.e, protons and deuterons) was determined by the data collected from the silicon telescope array. Data was filtered in the analysis code in order to isolate physically-relevant events of interest that occurred within the test chamber. For instance, electrical signals that did not correspond with a physically relevant trajectory through the particle telescope were thrown out. Particle identification was accomplished by creating a two dimensional matrix of energy deposited in the ΔE (front) detector vs. the energy deposited in the E (back) detector. This matrix generated two different regions representative of the different ejectiles present in the experiment.

These regions are referred to as “bananas” and can be seen in Fig. 3.6. The following plots correspond to the ^{92}Mo data set, however, a similar analysis was conducted for the ^{94}Mo data as well. In Fig. 3.6 the lower region corresponds to the proton ejectile with a resemblance closer to a “banana” shape while the relatively intense, upper region is representative of the deuteron, which lacks the “banana” shape due to the intensity of elastic scattering.

In order to determine the number of single particle proton events, $N_{(d,p)}$, a gate had to be incorporated in order to isolate protons at all energies. Due to the close proximity of the energies of the deuteron banana with the proton banana, illustrated in Fig. 3.6, a free form “banana” gate is difficult to accurately make and could lead to some of the proton data being misinterpreted. A more accurate method of particle

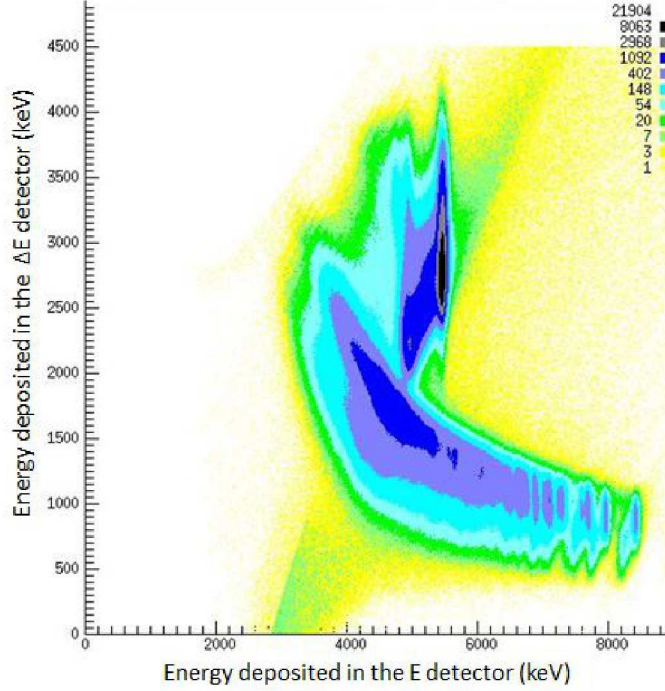


Figure 3.6: A “banana” plot from an analysis of the energy deposited in the ΔE (front) detector vs. the energy deposited in the E (back) detector for the $^{92}\text{Mo}(d, p)$ reaction.

identification lies within the linearization of the energies of the proton and deuteron ejectiles. A particle identification number (PID) linearization is given by Eq. 3.1.

$$\text{LINPID} = [(\Delta E + E)^{1.73} - E^{1.73}] \cos(\theta) \quad (3.1)$$

where ΔE is the energy deposited in the ΔE detector, E is the energy deposited in the E detector and θ is the polar angle of detection of the ejectile (as determined from the annular ring identification number discussed in Sec. 3.1.1). Figure 3.7 shows the plot obtained by incorporating the LINPID equation. Two distinct regions are made clearly distinguishable. The first region represents protons and the second represents deuterons. The shoulders, or small bumps are due to the coefficients used in Eq. 3.2.1. Gates were set at the solid lines in Fig. 3.7 and were employed in order to produce a new banana plot which consists of solely proton data shown in Fig. 3.8.

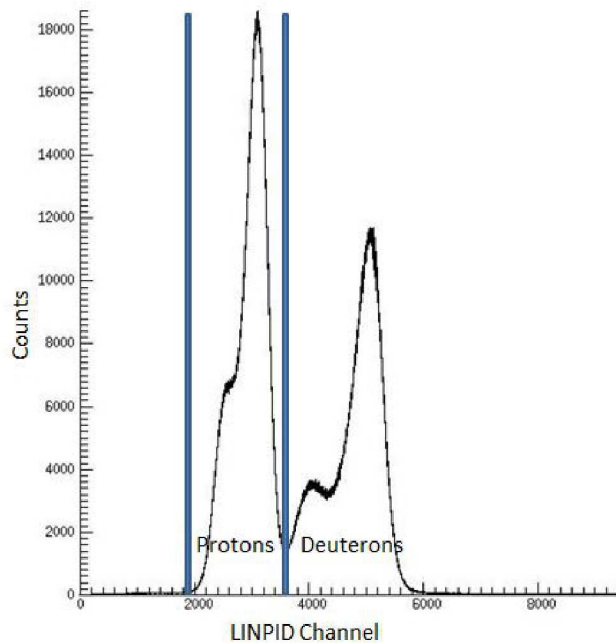


Figure 3.7: Linearized particle identification plot for the ^{92}Mo target data. A similar analysis was conducted using the ^{94}Mo data as well. “LINPID Channel” refers to the value calculated from Eq. 3.2.1.

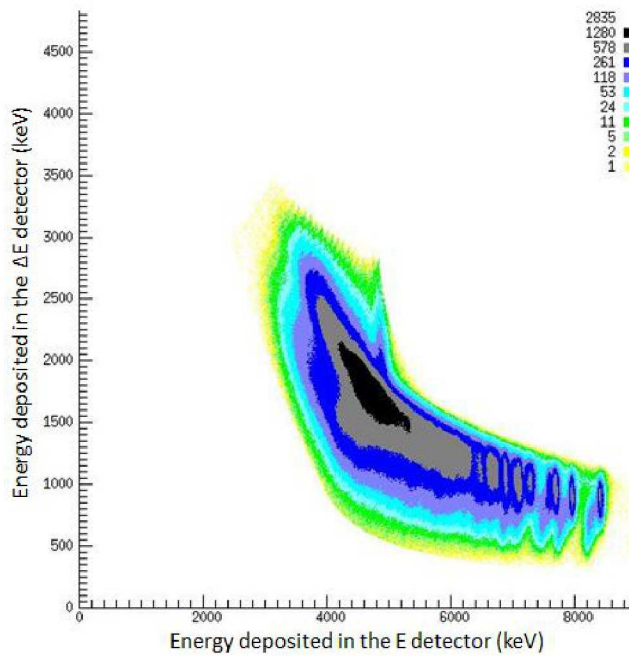


Figure 3.8: Proton-gated “banana” plot from an analysis of the energy deposited in the ΔE (front) detector vs. the energy deposited in the E (back) detector for the $^{92}\text{Mo}(d, p)$ reaction.

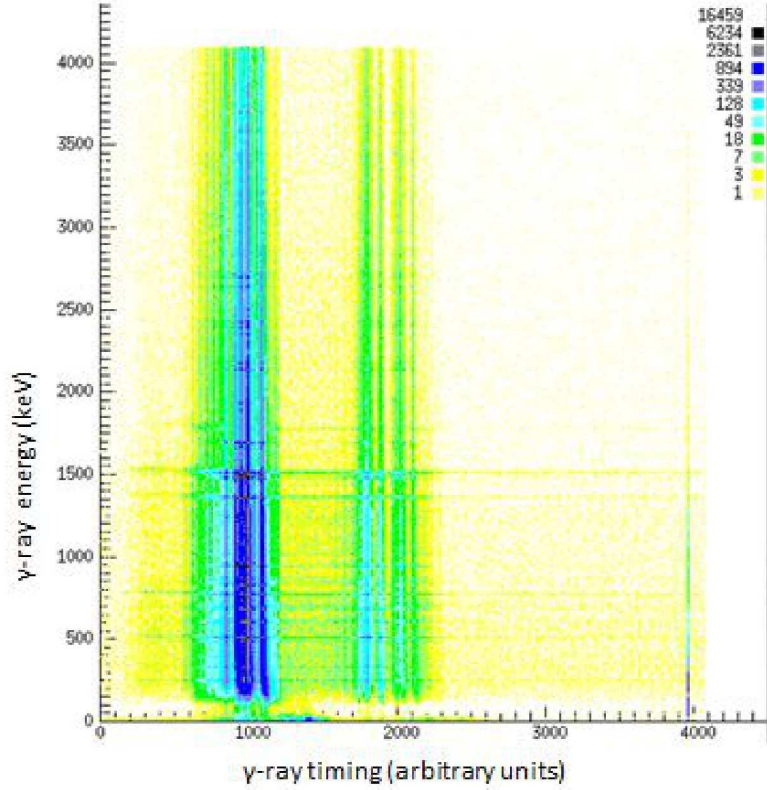


Figure 3.9: 2-D matrix of γ -ray energy versus γ -ray timing

3.2.2 Proton- γ Coincident Events

The next step in the analysis is to determine the number of γ -rays in coincidence with protons as a function of excitation energy in the compound nucleus for the surrogate measurement, $N_{(d,p\gamma)}(E_x)$. In order to do this timing spectra were utilized. A 2-D plot with γ -ray energy versus clover timing was generated shown in Fig. 3.9. The first high intensity region was taken as the prompt- γ coincident events. Using a timing gate on the first high intensity region in Fig. 3.9 protons in coincidence with γ events were determined, which is shown in comparison with non-coincident proton events plotted in Fig. 3.10. It is important to notice that the extreme right peak on the left plot is not present in the coincident plot on the right because a non-excited recoil nucleus does not produce γ -rays.

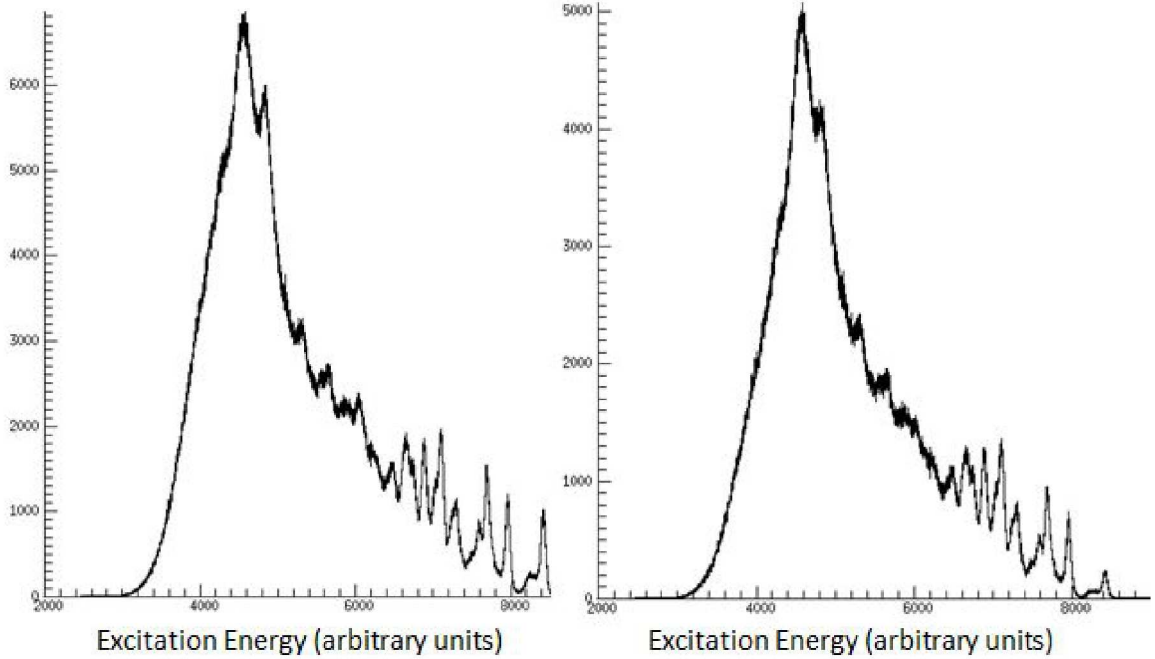


Figure 3.10: Left: Total number of (d, p) events. Right: Total number of (d, p) reactions in coincidence with γ -rays emitted from the compound nucleus.

3.2.3 Excitation Energy Calibration

The energy of the proton ejectile detected in the STARS array must be put in terms of excitation energy in the compound nucleus populated from the (d, p) reaction in order to generate cross sections. In Fig. 3.10, protons in coincidence with γ -rays showed resonances in the kinetic energy of the proton, which correspond to discrete states populated in the residual nucleus. Fig. 3.11 was generated by gating on the γ -ray energy of a decay from a specific energy level in the ^{93}Mo nucleus, which created a plot for protons in coincidence with that γ -ray energy.

For the case in Fig. 3.11, a γ -ray of 943 keV of energy is produced from an energy level at 943 keV in the excited nucleus. By gating on the 943 keV γ -ray emitted from an energy level of 943 keV a coincident proton spectrum was obtained, the resonance was fit for the centroid of the coincident proton. The centroid of the proton peak from the coincident 943 keV γ -ray gave a single data point for the calibration. By gating on several resonances in the compound nucleus and fitting each peak to determine the

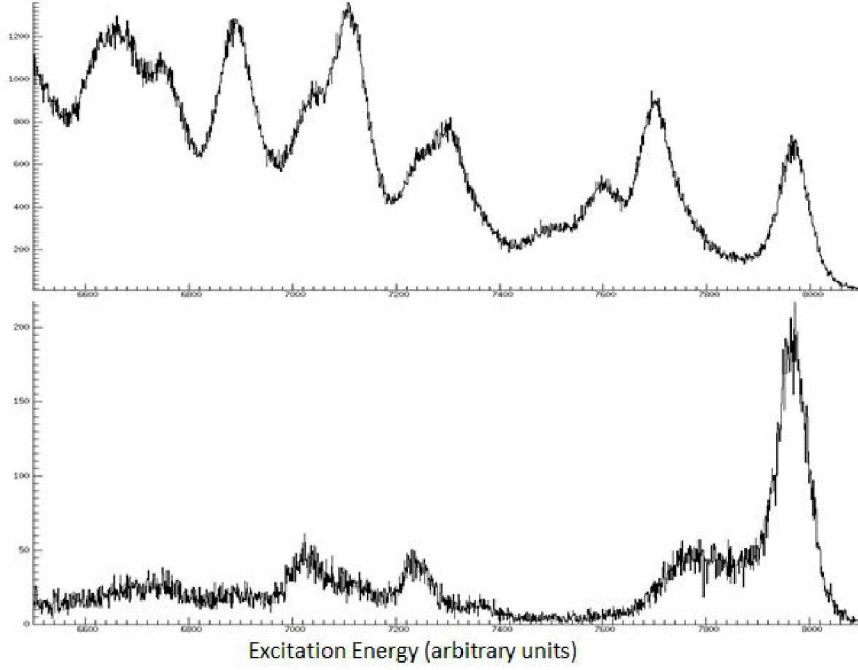


Figure 3.11: Top: Total coincident proton spectrum for the ^{93}Mo residual nucleus between channel 6500 and 8100. Bottom: Protons in coincidence with γ -rays emitted from the first excited state in the ^{93}Mo residual nucleus (943 keV).

proper corresponding centroid (in channels), a linear regression of excitation energy in the residual nucleus as a function of channel was made as shown in Fig. 3.12.

This procedure was also applied to the ^{94}Mo data to create a similar linear regression which represents the excitation energy of the residual nucleus as a function of channel input from the proton singles spectrum. The excitation energy of the residual nucleus in the surrogate reaction can be related to the incident neutron energy, E_n , of the surrogate reaction via Eq. 3.2.

$$E_n = \frac{A+1}{A}(E_x - S_n) \quad (3.2)$$

where S_n is the binding energy or neutron separation energy in the target nucleus. The factor $\frac{A+1}{A}$ represents the nuclear-recoil energy imparted in the neutron-induced reaction on a nucleus consisting of A nucleons. Cross sections were obtained at excitation energies above S_n in order for E_n to return a positive value. For most

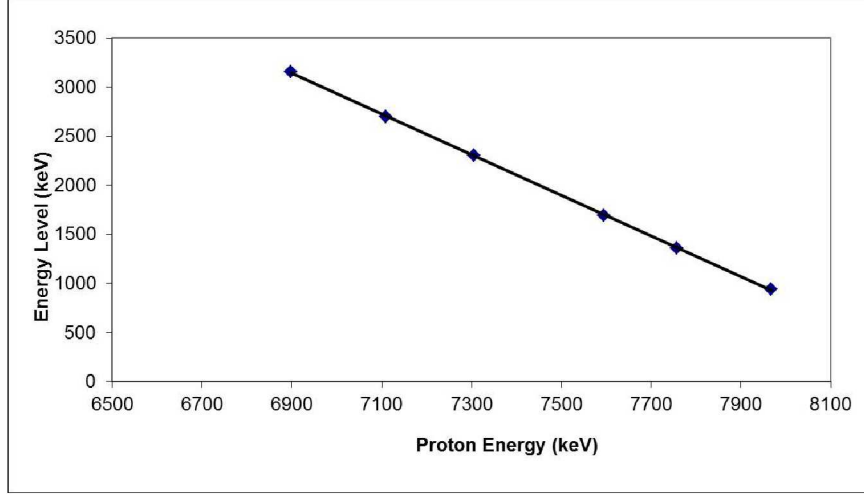


Figure 3.12: Linear regression using discrete states populated in the residual nucleus yielding an equation for excitation energy in the residual nucleus as a function of channel for the ^{92}Mo target data.

surrogate measurements, the data range is limited by the Coulomb barrier in the nucleus, however, in this case the data range was limited by apparently unphysical kinks in the cross section data given by ENDF for the ^{92}Mo and ^{94}Mo data. The kinks in the $^{92}\text{Mo}(n,\gamma)$ and $^{94}\text{Mo}(n,\gamma)$ cross section data are shown in Fig. 3.13 for reference [20]. For the $^{94}\text{Mo}(n,\gamma)$ cross section, the kink occurs at an energy of roughly 900 keV of incident neutron energy which limited the upper range for this surrogate measurement to 880 keV.

Finally, in order to find the number of γ -rays emitted from the residual nucleus created in the experiment, a two dimensional proton-gated, time-correlated matrix of proton-ejectile counts coincident with γ -rays as a function of excitation energy versus coincident γ -ray counts as a function of energy was obtained for both the $^{92}\text{Mo}(d,p)$ and $^{94}\text{Mo}(d,p)$ reactions, an example of which is shown in Fig. 3.14. The γ -ray spectra for γ -decay channel tagging measurements were created by using 100 keV energy gates on the ejectile as a function of excitation energy. This allowed for the extraction of γ -ray spectra in coincidence with the proton-ejectile and ensured that the γ -rays measured are from a particular energy range in the excited residual nucleus. These γ spectra were then background subtracted and efficiency corrected in order

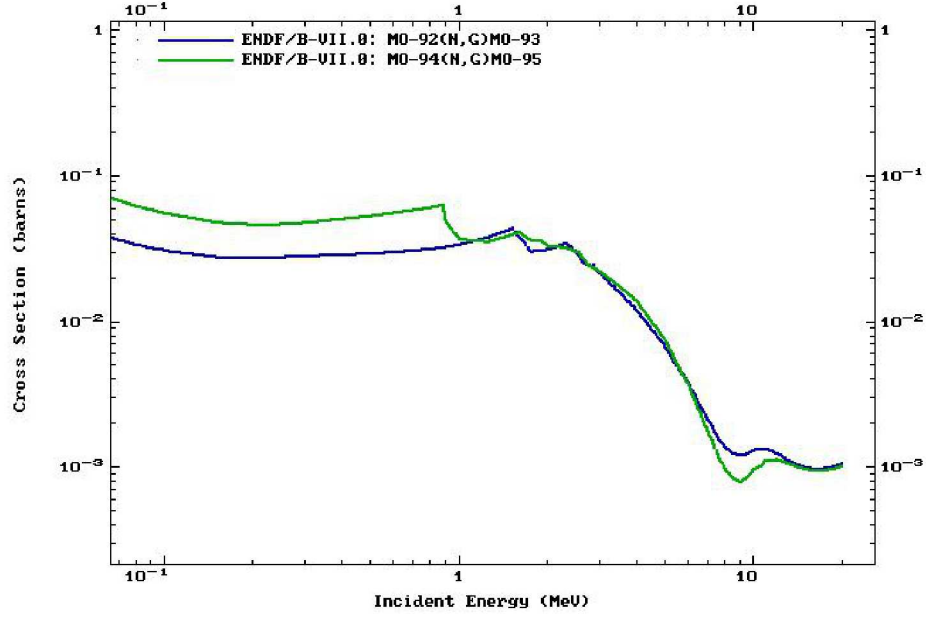


Figure 3.13: The $^{92}\text{Mo}(n,\gamma)$ (blue) and $^{94}\text{Mo}(n,\gamma)$ (green) cross section provided by ENDF.

Table 3.1: Isotopic Composition of the ^{92}Mo Target

Isotope	Atomic Percent	Precision
92	98.27	± 0.10
94	0.46	± 0.05
95	0.37	± 0.03
96	0.26	± 0.03
97	0.13	± 0.03
98	0.27	± 0.03
100	0.25	± 0.03

to apply the two γ -decay channel tagging techniques to obtain the surrogate cross section measurements.

3.3 Target Composition

Although the isotopic composition of the ^{92}Mo target was known, given in Table 3.1, a detailed composition of the ^{94}Mo was not readily available. In order to determine the isotopic composition of the ^{94}Mo target an analysis of γ -ray spectra was

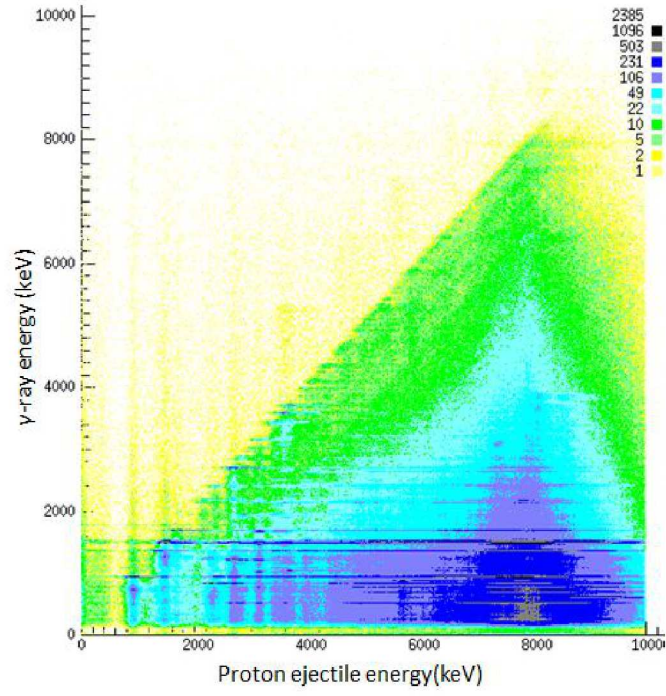


Figure 3.14: Two dimensional proton-gated, time-correlated matrix of proton-ejectile counts coincident with γ rays as a function of excitation energy versus coincident γ -ray counts as a function of energy

conducted. These γ -ray spectra were generated by gating on the deuteron inelastic scattering channel in order to isolate γ -rays in coincidence with inelastically scattered deuterons. The prominent γ -rays detected were corrected for detector efficiency and then referenced in the NNDC library for contributions from Mo isotopes, which led to the generation of a list of isotopic possible contaminants in the target [28]. Only stable isotopes were considered. Through an analysis of the γ -ray spectra for states with similar angular momentum and transitions with significant intensities, the list of possible isotopic contaminants was narrowed down to only a few isotopic contaminants: ^{92}Mo , ^{95}Mo , and ^{100}Mo . By fitting the peak areas and determining a pure ^{94}Mo peak in the spectrum at 869 keV, relative weights were assigned to the isotopic contaminants γ -ray peaks. The relative weights of the isotopic contaminate peaks served as an estimate of the individual contributions of these isotopes to the target composition. These isotopes contributed roughly 5% to the total target composition which yielded a 95% pure ^{94}Mo target.

Chapter 4

Results and Discussion

As concluded in Ch. 3 a two-dimensional, proton-gated matrix of γ -ray energy and compound nuclear excitation energy, E_x , as determined by the proton energy, corrected for energy losses, was obtained for both the $^{92}\text{Mo}(d,p)$ and $^{94}\text{Mo}(d,p)$ reactions. For each excitation energy bin in the compound nucleus, γ -ray spectra covering an energy range of approximately 100 keV to 8 MeV were recorded and each γ -ray spectrum was divided by the γ -detection efficiency of the HPGe array. Both the statistical and discrete γ -decay channel tagging techniques were applied to the extracted γ -ray spectra in order to generate cross sections using the ASM and SRM.

4.1 Absolute Surrogate Measurement

Figure 4.1 shows the $^{92}\text{Mo}(n,\gamma)$ cross section obtained via the absolute surrogate method as described in Eq. 2.1 as a function of equivalent neutron energy. Both the discrete (open circles) and statistical (filled squares) γ -decay tagging techniques were employed. The shaded region represents the evaluated $^{92}\text{Mo}(n,\gamma)$ cross section obtained from ENDF/B-VII.0 with associated 18% uncertainty [29]. It is important to note that no experimental data exists above 125 keV range to support the $^{92}\text{Mo}(n,\gamma)$ cross section, rather the evaluated data above this range is calculation based.

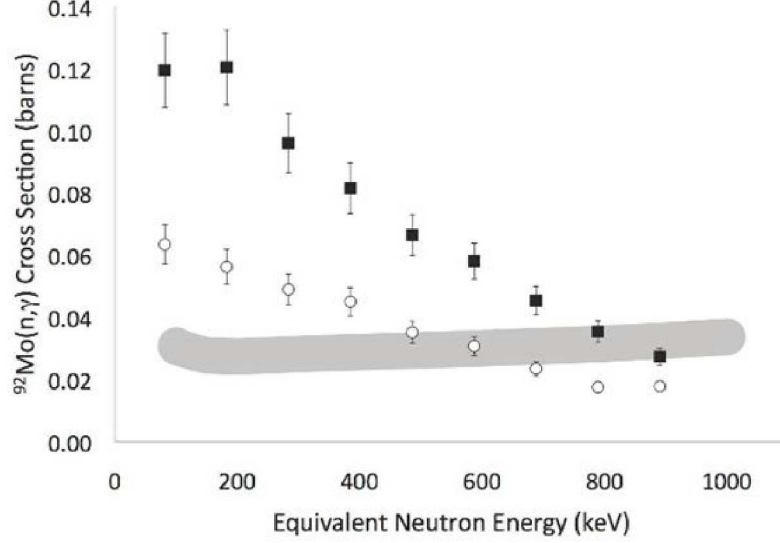


Figure 4.1: The absolute surrogate $^{92}\text{Mo}(n,\gamma)$ cross section obtained via the discrete (open circles) and statistical (filled squares) γ -decay tagging techniques as a function of equivalent neutron energy from the (d,p) surrogate reaction is displayed above. The error bars represent both the statistical and non statistical uncertainty. The evaluated $^{92}\text{Mo}(n,\gamma)$ cross section from ENDF/B-VII.0 with associated uncertainty denoted by the shaded region is shown for comparison.

The absolute surrogate cross section obtained using the discrete γ -decay channel tagging technique shows a closer agreement with the ENDF/B-VII evaluation than the statistical γ -decay tagging technique over the equivalent neutron energy range with an isolated agreement with the data between 480 keV and 680 keV. However, the exhibited shape of discrete γ decay tag results does not trend well with the evaluated result. Using the statistical γ -decay tagging technique to measure the absolute surrogate cross section yielded results that deviated from the ENDF/B-VII.0 evaluation by as much as a factor of 4 at lower equivalent neutron energies and also exhibited an inconsistent shape with the evaluated data with an isolated agreement between 790 and 890 keV. The discrepancies between the surrogate and evaluated data may imply a failure of the Weisskopf-Ewing approximation required to obtain Eq. 2.1 and/or that the tagging techniques do not serve as a robust tag of the compound nucleus.

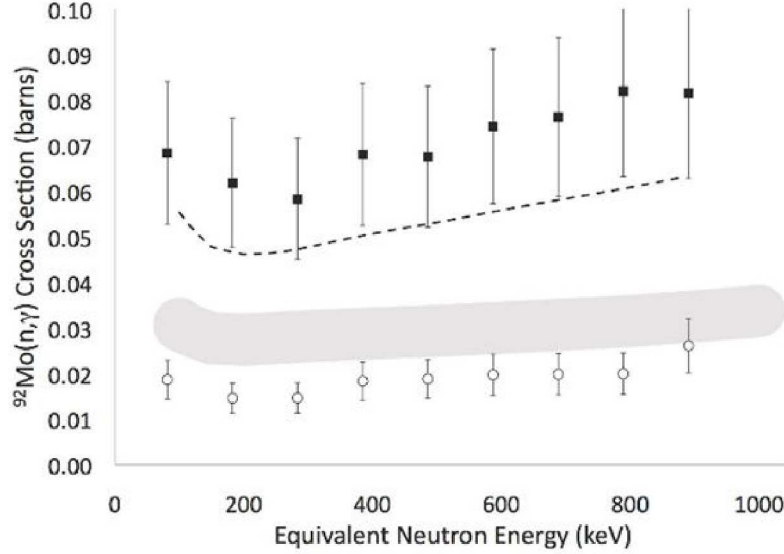


Figure 4.2: The $^{92}\text{Mo}(n,\gamma)$ cross section obtained via the SRM using the discrete (open circles) and statistical (filled squares) γ -decay tagging techniques as a function of equivalent neutron energy obtained via the (d,p) surrogate reaction. The error bars represent both the statistical and non-statistical uncertainty. The evaluated $^{92}\text{Mo}(n,\gamma)$ cross section from ENDF/B-VII.0 with associated uncertainty denoted by the shaded region is shown for comparison. The evaluated $^{94}\text{Mo}(n,\gamma)$ cross section from ENDF/B-VII.0 is shown by the dashed line.

4.2 Surrogate Ratio Measurement

The surrogate ratio measurement was determined by applying the deuteron beam on both the ^{92}Mo and ^{94}Mo targets. Figure 4.2 shows the $^{92}\text{Mo}(n,\gamma)$ cross section obtained via the SRM using the discrete (open circles) and statistical (filled squares) γ -decay tagging techniques as a function of equivalent neutron energy obtained via the (d,p) surrogate reaction. The error bars represent both the statistical and non-statistical uncertainty. The evaluated $^{92}\text{Mo}(n,\gamma)$ cross section from ENDF/B-VII.0 with associated uncertainty denoted by the shaded region is shown for comparison. The evaluated $^{94}\text{Mo}(n,\gamma)$ cross section from ENDF/B-VII.0 is shown by the dashed line.

The SRM $^{92}\text{Mo}(n,\gamma)$ cross section obtained through the employment of the discrete γ -decay tagging technique shows a similar shape as the ENDF/B.VII. 0 evaluation,

but is systematically lower by a factor of roughly 1.5 to 2 over the range of equivalent neutron energies selected (taking note of the singular agreement of the data point at 890 keV). The discrepancy between the evaluated and measured data, using the discrete γ -decay tagging technique, may lie in the initial spin mismatch in the transitions used. For the ^{93}Mo residual nucleus the lowest transition is at 943.28 keV ($1/2^+ \rightarrow 5/2^+$) whereas the ^{95}Mo residual nucleus used the 204.12 keV ($3/2^+ \rightarrow 5/2^+$) transition. This initial angular momentum mismatch may introduce uncorrelated errors in the measurement if the assumption of angular momentum independence in the Weisskopf-Ewing approximation is not applicable. An ideal surrogate ratio measurement would utilize transitions with very similar nuclear structure and energy in the discrete γ -decay tag. It is interesting to note that, within the Weisskopf-Ewing limit, the greatest deviations from the evaluated data are expected to occur at lower equivalent neutron energy and the surrogate data would be expected to converge with the evaluated data as equivalent neutron energy increases.

The surrogate $^{92}\text{Mo}(n,\gamma)$ cross section obtained using the statistical γ -decay tagging technique shows no agreement with the ENDF/B.VII. 0 evaluation within estimated uncertainty over the entire equivalent neutron energy range probed. The surrogate $^{92}\text{Mo}(n,\gamma)$ cross section obtained using the statistical γ -decay tagging technique is systematically higher than the evaluation by a factor of 2 to 3 over the entire range and trends well with the $^{94}\text{Mo}(n,\gamma)$ fiducial cross section, suggesting that the ratio of decay probabilities scales poorly towards the desired cross section.

Despite the disparities discussed above, cross sections derived using the SRM provided closer agreement with the $^{92}\text{Mo}(n,\gamma)$ evaluated data than the absolute surrogate method (note the scale of Fig. 4.2). This suggests that possible correlated errors such as effects of the angular momentum population distribution, neutron leakage and effects from the Compton continuum, and similar errors in the experimental set-up cancel in the ratio analysis due to the similar nature of the two nuclei involved. These errors were previously unaccounted for in the absolute surrogate measurement.

4.3 Evaluation of γ -decay Tagging Techniques

In order to understand the differences in the measured cross section results through the employment of the discrete and statistical γ -decay tagging techniques an analysis of the γ -decay probabilities of the ^{93}Mo and ^{95}Mo residual nuclei was conducted. The γ -decay probabilities were corrected for contributions from the Compton continuum by subtracting the average Compton contributions from the energies relevant for the γ -decay probability. As illustrated in Fig. 4.3, the γ -decay probability of the ^{93}Mo (filled symbols) and ^{95}Mo (open symbols) residual nuclei was obtained using the discrete and statistical γ -decay techniques as a function of excitation energy in the residual nuclei. The γ -decay probabilities obtained using the statistical γ -decay tagging technique (squares) differ from the γ -decay probabilities obtained using the discrete γ -decay tagging technique (circles) by roughly a factor of 2 at energies above separation energy of the ^{93}Mo residual nucleus, indicating that the two methods for tagging the γ -decay channel capture different information.

The dashed line in Fig. 2.2 represents the neutron separation energy in the ^{93}Mo residual nucleus ($S_n = 8.06981$ MeV), whereas the dotted line represents the neutron separation energy of the ^{95}Mo residual nucleus ($S_n = 7.3691$ MeV). As the excitation energy in the ^{93}Mo and ^{95}Mo residual nuclei increases beyond the neutron separation energy, the γ -decay probabilities are expected to decrease with the opening of the neutron emission channel. Evidence of this decrease in γ -decay probabilities are shown by the sharp decrease across the $S_n = 7.3691$ MeV line in the ^{95}Mo residual nucleus (filled markers) and the rather smooth decrease across the $S_n = 8.06981$ MeV in the ^{93}Mo residual nucleus (open markers).

The γ -decay probabilities below the neutron separation energy are expected to be unity due to γ -decay being the only open decay channel, as neutron emission is highly improbable below the neutron separation energy. Figure 2.2 shows that both discrete and statistical tagging techniques have measured γ -decay probabilities for the ^{93}Mo

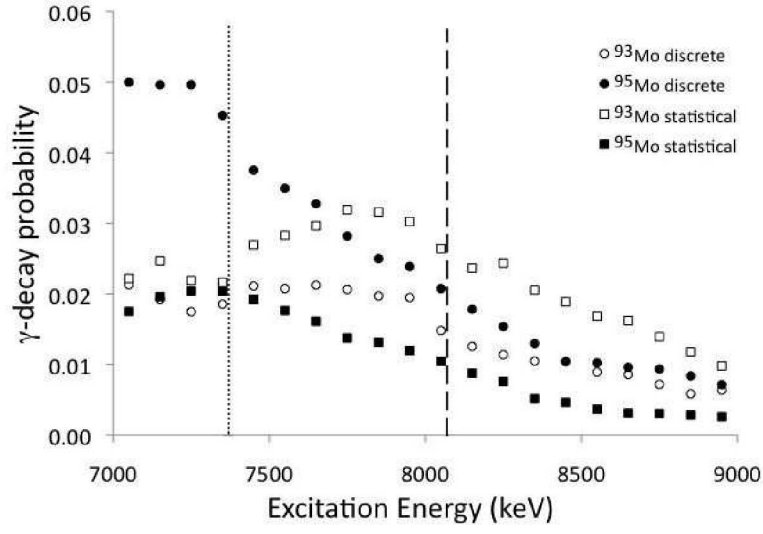


Figure 4.3: The γ -decay probability of the ^{93}Mo (open symbols) and ^{95}Mo (filled symbols) residual nuclei obtained using the discrete (circles) and statistical (squares) γ -decay techniques as a function of excitation energy in the residual nucleus. The dashed line represents the neutron separation energy of the ^{93}Mo residual nucleus ($S_n = 8.06981$ MeV). The dotted line represents the neutron separation energy of the ^{95}Mo residual nucleus ($S_n = 7.3691$ MeV). The statistical error is smaller than the data points.

and ^{95}Mo residual nuclei much less than unity below the neutron separation energy, indicating incomplete collection of the γ -decay channel for both tagging techniques.

The discrete method relies on the assumption that all γ -cascades pass through the transition of interest. The transitions used were at 943.28 keV ($1/2^+ \rightarrow 5/2^+$) in the ^{93}Mo residual nucleus and at 204.12 keV ($3/2^+ \rightarrow 5/2^+$) in the ^{95}Mo residual nucleus. The fraction of cascades that pass through these transitions are unknown and may be less than unity, which may introduce an error in the measured γ -decay probability. Previous work employed a correction factor for cascades that did not pass through the transitions of interest [15].

Further, in order to determine the information captured using the two different tagging techniques a ratio was employed which related the number of particle- γ coincident events tagged using the statistical tagging technique $N_{d,p\gamma}^{stat}(E_x)$ to the number of particle- γ -coincident events tagged using the discrete tagging technique displayed in Eq. 4.1:

$$R = \frac{N_{d,p\gamma}^{stat}(E_x)}{N_{d,p\gamma}^{disc}(E_x)} \quad (4.1)$$

If these two tagging techniques capture the same information over the entire excitation energy range than this ratio should remain constant over the entire energy range. If either tagging method is significantly influenced by the initial angular momentum population distribution, it is unlikely that a constant value will be obtained for this ratio as a function of excitation energy.

Figure 4.4 illustrates the results of the ratio in Eq. 4.1 obtained over the 7-9 MeV energy range for the ^{93}Mo (filled triangles) and ^{95}Mo (open diamonds) residual nuclei. Over the entire sampled excitation energy range, the ^{95}Mo residual nucleus showed limited variability in the ratio and seemingly approached a constant value at roughly 0.5 as excitation energy increased. It appears that angular momentum effects are less notable for the relatively more deformed ^{95}Mo residual nucleus and both the discrete and statistical tag capture similar information. The ^{93}Mo residual

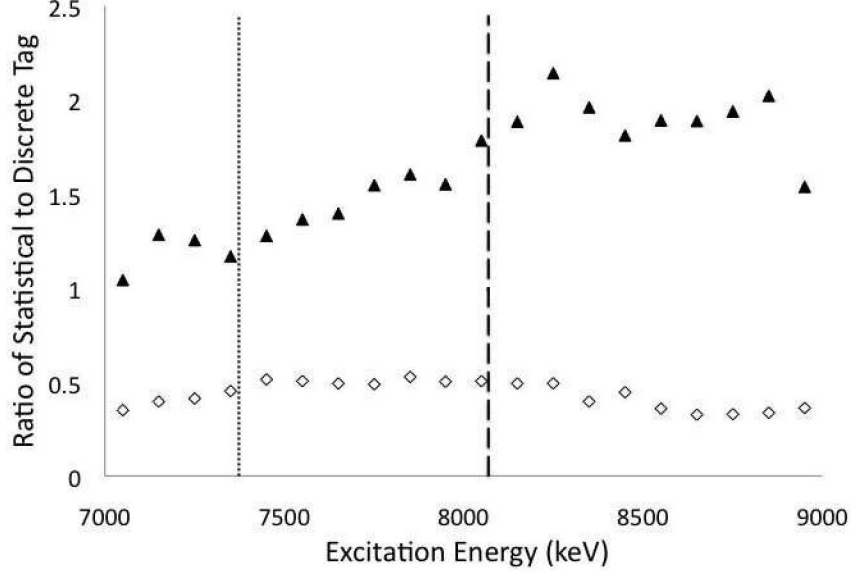


Figure 4.4: Ratio of the statistical to discrete γ -decay tags as a function of excitation energy in the ^{93}Mo (filled triangles) and ^{95}Mo (open diamonds) residual nuclei. The dashed line represents the neutron separation energy in the ^{93}Mo compound nucleus. The dotted line represents the neutron separation energy in the ^{95}Mo compound nucleus. In some cases, the statistical error is smaller than the data points.

nucleus data shows an increased variability in the ratio which becomes even more sporadic above the neutron separation energy. This variability may be accounted for by an increased sensitivity to the angular momentum population distributions in the ^{93}Mo residual nucleus due to its proximity to the $N = 50$ neutron shell enclosure. The ^{93}Mo residual nucleus is more spherical in nature, due to its close proximity to the $N = 50$ neutron shell enclosure, which results in a less dense low energy level density in comparison to the more deformed ^{95}Mo residual nucleus. The Weisskopf-Ewing approximation requires that the level density be sufficiently dense in order to be represented by a continuous function. A less dense distribution low-energy level density in the spherical ^{93}Mo residual nucleus may result in the breakdown of the Weisskopf-Ewing approximation and a greater dependence in the angular momentum distribution in the populated ^{93}Mo residual nucleus.

Chapter 5

Conclusions

The indirect measurement of the $^{92}\text{Mo}(n,\gamma)$ cross section via the surrogate method using two different γ -decay channel techniques, discrete and statistical, was determined over an energy range of 80 keV to 890 keV, highlighting the first surrogate measurement on spherical and near-spherical nuclei in the mass-90 region. The data extracted via the absolute surrogate measurement using both the statistical and discrete γ -decay tagging techniques showed poor agreement with the evaluated data. Cross sections extracted via the SRM showed a decrease in correlated errors for both employed γ -decay tagging techniques.

In order to improve measurements in $^{92}\text{Mo}(n,\gamma)$ surrogate cross sections using the discrete tagging technique, empirical data could be employed to better understand the γ -decay cascades through the transitions of interest for the relevant nuclei and potentially sum parallel γ -decay paths. Improvements in the statistical tagging technique can be made by using calculations and model simulations in order to constrain the γ -ray energy gate to isolate primary γ -transitions. An evaluation of the γ -decay tagging techniques indicated that these techniques are more likely to capture similar information in the more deformed ^{95}Mo residual nucleus as opposed to more variations in information collected in the quasi-spherical ^{93}Mo residual nucleus. Future work should include an extension of the equivalent neutron energy range above

1 MeV to analyze the rapid changes in the $^{93}\text{Mo}(n, \gamma)$ evaluated cross section, as well as a comprehensive theoretical evaluation of the statistical and discrete γ -decay tagging techniques.

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