Statistical γ rays in the analysis of surrogate nuclear reactions

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The surrogate nuclear reaction method is being applied in many efforts to indirectly determine neutron-induced reaction cross sections on short-lived isotopes. This technique aims to extract accurate (n,γ) cross sections from measured decay properties of the compound nucleus of interest (created using a different reaction). The advantages and limitations of a method that identifies the γ-ray decay channel by detecting any high-energy ("statistical") γ ray emitted during the relaxation of the compound nucleus were investigated. Data collected using the Silicon Telescope Array for Reaction Studies and Livermore-Berkeley Array for Collaborative Experiments silicon and germanium detector arrays were used to study the decay of excited gadolinium nuclei following inelastic proton scattering. In many cases, this method of identifying the γ-ray decay channel can simplify the experimental data collection and greatly improve the detection efficiency for γ-ray cascades. The results show sensitivity to angular-momentum differences between the surrogate reaction and the desired (n,γ) reaction similar to an analysis performed using low-lying discrete transitions even when ratios of cross sections are considered.

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I. INTRODUCTION

Radiative neutron-capture reactions on unstable nuclei play important roles in many areas of basic and applied science. The competition between radiative neutron capture and β-decay processes for many nuclei with half-lives ranging from weeks to years influences the synthesis of heavy elements by the astrophysical s process [1]. Cross sections for these nuclei are required to uncover the properties of the s-process environments from observed isotopic abundances [2]. Evaluations of generation IV nuclear reactor designs and novel fuel-cycle concepts require neutron-capture cross sections on actinides and some of the longer-lived fission products [3,4]. In addition, accurate (n,γ) cross-section measurements are needed for a variety of applications in homeland security and stockpile stewardship.

Direct measurements of (n,γ) cross sections for radioactive nuclei are extremely challenging because of the target activity and the difficulty in collecting the required quantity of material. To date, calorimetric measurements performed using the high-efficiency, highly segmented detector arrays of the CERN neutron time-of-flight facility (n_TOF) [5] and the Detector for Advanced Neutron Capture Experiments (DANCE) [6] have reached a precision of 2–10% for isotopes with half-lives as short as ∼100 years [7–9]. This type of measurement becomes increasingly more difficult for isotopes with shorter half-lives. For reactions where the capture product is also a radioactive nucleus, activation techniques can be performed on smaller quantities of target material and therefore can be used to study shorter-lived samples (for recent measurements on short-lived and sub-microgram-quantity samples, see Refs. [10,11], respectively). However, neutron capture on most s-process branching-point nuclei and many other isotopes of interest results in stable nuclei.

The surrogate nuclear reaction method [12,13] has received significant attention recently [14] as a viable indirect approach to determine (n,γ) cross sections on short-lived or extremely rare isotopes. The technique has successfully been shown to determine (n, f) cross sections for nuclei within several nucleons of a stable or long-lived isotope [15–17].

Cross sections for two-step nuclear reactions that proceed through a highly excited, equilibrated compound nucleus can be determined by combining reaction-model results with measured decay properties of the compound nucleus [12,18]. Hauser-Feshbach theory [19] is used to express an (n,γ) cross section, $\sigma_{n\gamma}(E_n)$, in terms of the cross section $\sigma_n^{\rm CN}(E_n, J, \pi)$ for the formation of the compound nucleus with spin parity J^π at neutron energy E_n and the exit-channel branching ratio $G_\gamma^{\rm CN}(E_{\rm ex}, J, \pi)$ for γ-ray decay as

$$\sigma_{n\gamma}(E_n) = \sum_{J,\pi} \sigma_n^{\rm CN}(E_n, J, \pi) G_\gamma^{\rm CN}(E_{\rm ex}, J, \pi). \quad (1)$$

Here width-fluctuation correlations between incident and outgoing channels are neglected. The excitation energy of the decaying compound nucleus, $E_{\rm ex}$, is related to the neutron energy, E_n, via

$$E_n = \frac{A+1}{A}(E_{\rm ex} - S_n), \quad (2)$$

where the near-unity factor $(A+1)/A$ accounts for the nuclear recoil imparted in the neutron-induced reaction on a nucleus consisting of A nucleons. The $\sigma_n^{\rm CN}$ cross sections can, in many cases, be accurately determined using optical models. The $G_\gamma^{\rm CN}$ exit-channel probabilities, however, are typically difficult to calculate reliably and therefore guidance must be provided from experimental studies. The goal of the surrogate nuclear reaction approach is to determine $G_\gamma^{\rm CN}$ indirectly by producing the desired compound nucleus at the energies of interest (albeit likely with a different J^π distribution) using an alternative (or "surrogate") reaction. This surrogate reaction is chosen to simplify the data collection and so typically involves a *light-ion* reaction on a *stable* (or more readily accessible) target.

Initial attempts to apply the surrogate nuclear reaction technique [20–24] to determine (n,γ) cross sections have come across two major challenges. First, for most reactions the compound-nuclear spin-parity (J^π) distributions are "mismatched"—the distributions produced in the desired and surrogate reactions are different. Experimental details such as the angular coverage of the light-ion detectors or the particular signature used to identify the γ-ray cascade will, in general, also have an impact on which subset of the overall compound-nuclear J^π distribution produced in the surrogate reaction is detected. Neutron-capture reactions are expected to be sensitive to spin-parity differences [13,25], especially for isotopes near a closed nucleon shell [18], where the nuclear level density is low. For (n,f) reactions these effects arise at low energies [12] but seem to have minimal impact on cross-section determinations at energies $\gtrsim$1 MeV [15,16]. Second, even for the best cases, collecting the desired statistics above S_n for the γ-ray exit channel is difficult because neutron (and potentially other particle) emission rapidly becomes the dominant decay channel [20,21,26]. In addition, in many cases, no single γ ray is emitted in a large fraction of cascades due to a lack of a strong low-lying collector transition or the presence of many highly converted transitions (such as in high-Z nuclei). This provides an additional challenge for techniques that seek to determine the exit channel by identifying discrete γ rays from the compound nucleus.

The difficulties associated with collecting statistics can be circumvented by identifying the γ-ray exit channel by detecting any γ ray emitted by the compound nucleus. Detector arrays have been used to efficiently identify the exit channel from any energy deposition above a certain threshold [22,23] (as opposed to using discrete lines as in Refs. [20,21]). As there are a large number of states at excitation energies above S_n (as well as a large number of energetically accessible states to decay to), the γ-ray emission spectrum can be very broad. These γ rays are both the high-energy γ rays emitted by the compound nucleus during the initial (or near the initial) step of the cascade as well as higher-energy transitions near the bottom of the cascade. These γ rays have been referred to as "statistical" γ rays in the literature [22,23], and this terminology is used here to describe any γ ray that deposits energy above a selected energy threshold, $E_{\rm th}$, in a detector. Ultimately, nearly 100% efficiency could be obtained using a 4π calorimeter detector, such as DANCE [6].

In addition, following some recent experimental work in which statistical γ rays were used to identify the exit channel, it has been suggested that the angular-momentum issues can be greatly reduced by determining ratios of (n,γ) cross sections from ratios of the decay channels for two compound nuclei with similar structures [22,23]. This would be a fortuitous simplification of the surrogate nuclear reaction technique. The reaction theory needed to interpret the decay data as an (n,γ) cross section could take advantage of the Weisskopf-Ewing limit of the Hauser-Feshbach theory [27] that has worked so well for (n,f) reactions. In this approximation, the branching ratios $G_\gamma^{\rm CN}$ are independent of J^π and Eq. (1) simplifies to

$$\sigma_{n\gamma}(E_n) = \sigma_n^{\rm CN}(E_n) G_\gamma^{\rm CN}(E_{\rm ex}). \quad (3)$$

Here the exit-channel probability measured in the surrogate reaction determines $G_\gamma^{\rm CN}$ directly and the compound-nucleus formation cross section, $\sigma_\gamma^{\rm CN}$, can be calculated using an optical model.

In this surrogate ratio approach [12,28], two surrogate experiments are performed to determine the ratio of two cross sections of compound-nuclear reactions. An independent determination of one cross section can then be used to deduce the other from the ratio. In this limit, the ratio $R(E_n)$ of the (n,γ) cross sections is

$$R(E_n) = \frac{\sigma_n^{\rm CN1}(E_n) G_\gamma^{\rm CN1}(E_n)}{\sigma_n^{\rm CN2}(E_n) G_\gamma^{\rm CN2}(E_n)}. \quad (4)$$

Often, it is assumed that $\sigma_n^{\rm CN1}/\sigma_n^{\rm CN2} \approx 1$ and therefore the ratio is determined solely from the ratio of the experimentally determined exit-channel probabilities. However, experiments that have determined (n,γ) cross sections from ratios of exit channels determined from discrete γ-ray transitions in Gd [20] and Yb [21] isotopes have shown sensitivity to angular-momentum effects consistent with the calculations in Ref. [25] and yield results that are a factor of $\sim$2 different from direct measurement.

In this paper, the use of a simple method to identify the γ-ray exit channel from any γ-ray signal above a threshold energy is explored. The results of a surrogate nuclear reaction analysis based on this signature are compared to the results previously obtained with low-lying discrete γ rays [20]. The advantages and limitations of this "statistical" γ-ray technique for determining cross sections is investigated.

II. EXPERIMENT

The data used here were collected from inelastic proton scattering to determine the exit-channel branching ratio for several gadolinium compound nuclei from the discrete ground-state band transition γ rays [20,29]. The gadolinium region is well suited for tests of the surrogate nuclear reaction method because many stable Gd isotopes exist for which (n,γ) cross sections have been directly measured (in some cases up to 1.0 MeV with quoted uncertainties below 1% [30] and up to 2.5 MeV with uncertainties of $\approx$10% [31]), and sufficient nuclear structure information is available to carry out complementary cross-section calculations. In addition, the angular-momentum considerations in reactions on these isotopes are expected to be nearly identical because the ground-state J^π of ^{155,157}Gd and ^{156,158}Gd are both $3/2^-$ and 0^+, respectively.

In this work, the branching ratio is determined from any signal in the γ-ray detectors above a selected energy, $E_{\rm th}$. These signals, after accounting for known backgrounds, can be attributed to γ rays emitted from the highly excited compound nucleus and many are likely to be statistical γ rays emitted during the first step of the γ-ray cascade. The experimental setup, detector calibration, and data analysis were described at length in Ref. [20] and are only summarized here. The handling of the γ-ray signals is different, however, and the impact on the data analysis and results is discussed in detail.

Isotopically enriched, $\approx$1 mg/cm^2 thick, self-supporting metal ^{154,155,156,158}Gd targets (enriched to 66.53%, 91.74%, 93.79%, and 92.00% respectively) were bombarded with $\approx$2 nA of protons with beam energy $E_b = 21.70 \pm 0.05$ MeV from the 88-Inch Cyclotron at the Lawrence Berkeley National Laboratory. Inelastic proton scattering served as the surrogate reaction for neutron capture. Proton-singles events (N_p) and proton-γ coincidence events ($N_{p\gamma}$) were collected using the Silicon Telescope Array for Reaction Studies (STARS) and five "clover" high-purity germanium (HPGe) detectors [32] with bismuth-germanate-oxide (BGO) Compton-suppression shields [33] of the Livermore-Berkeley Array for Collaborative Experiments (LiBerACE) [20,34]. The STARS detectors were arranged in a ΔE-$E1$-$E2$ telescope configuration consisting of double-sided silicon $S2$ detectors [35] of thicknesses 500 μm, 1000 μm, and 1000 μm. The ΔE detector was segmented into 48 rings (0.5 mm wide) and 16 sectors (each spanning 22.5°). The $E1$ and $E2$ detectors were operated with a 2× coarser segmentation. A 200 μg/cm^2 thick aluminum foil in front of the silicon stopped low-energy δ electrons that emerged from the target. A cooled copper heat shield surrounded the detector array and lowered the detector temperature to 15°C to reduce thermal noise.

The particle energy calibration was based on data collected offline with a ^{226}Ra α source and from scattering from low-lying discrete states in ^{12}C and ^{16}O collected online. Protons of energy 10–17 MeV (corresponding to excitation energies of 5–12 MeV) could be identified by their energy loss in the ΔE and $E1$ detectors [36]. The nuclear excitation energy was determined from $E_{\rm ex} = E_b - E_p - E_r$ where the scattered proton energy, E_p, was reconstructed with an uncertainty of 50 keV over the entire energy range and the kinetic energy imparted to struck Gd nuclei, E_r, was calculated event by event from the scattered proton kinematics. The 1-σ width of the elastic peak was determined to be 65 keV.

III. RESULTS

The probability that the nucleus de-excites by γ-ray emission, $P_{p\gamma}$, was determined from the relation

$$P_{p\gamma}(E_{\rm ex}) = \frac{1}{\epsilon_\gamma} \times \frac{N_{p\gamma}(E_{\rm ex})}{N_p(E_{\rm ex})}, \qquad (5)$$

where ϵ_γ is the efficiency for identifying the γ-ray cascade branch (which depends on $E_{\rm th}$ and nucleus) and the proton detection efficiency cancels in the ratio. N_p was determined in Ref. [20] for ^{156,158}Gd after correcting for carbon and oxygen contamination and the contribution from other Gd isotopes present due to imperfect enrichment.

The number of proton-γ-ray coincidences in which the total γ-ray energy deposited in the LiBerACE HPGe array was above $E_{\rm th}$ was used to determine $N_{p\gamma}$. The analysis was performed with $E_{\rm th} = 500$, 1000, 1500, 2000, and 2500 keV to explore the sensitivity of the results to the criteria used to identify a statistical γ ray. The results for $E_{\rm th} = 500$ keV and 2500 keV are shown in Fig. 1. Using a larger value for $E_{\rm th}$ is seen to increase the relative contribution from ^{12}C and ^{16}O background peaks because the first several excited states of these nuclei decay by high-energy γ-ray emission. This background structure was subtracted using the known peak shapes determined from N_p in Ref. [20] (the broader continuum under the peaks can be attributed to broad states that decay by particle emission which contribute to N_p but not $N_{p\gamma}$). The relative amplitudes are consistent with the known contamination of these targets. The uncertainty in the peak amplitudes was estimated to be ±7%. The small contribution to $N_{p\gamma}$ from the other Gd isotopes was subtracted off using the measured data (where possible) or estimates of the contribution based on data.

At excitation energies $E_{\rm ex} > S_n + E_{\rm th}$, an additional background is seen to arise from γ rays emitted from (p, pn) reactions that result in a different compound nucleus (these reactions would serve as the neutron-emission exit channel and correspond to the surrogate reaction for the (n, n') reaction). Therefore, the results for $P_{p\gamma}$ at these energies become unreliable. Higher values for $E_{\rm th}$ therefore provide a larger energy window that is free of γ-ray backgrounds from particle emission.

The efficiency ϵ_γ in Eq. (5) is, in practice, difficult to determine *a priori* because it depends on the details of complicated γ-ray emission spectra and the detector response to these spectra. Instead, ϵ_γ is determined from the constraint that $P_{p\gamma} = 1$ below the neutron separation energy because γ-ray emission is the only open decay channel. Indeed, each $P_{p\gamma}$ spectrum appears to reach a constant value at energies just below S_n, and ϵ_γ is chosen to make this value unity to satisfy the previously mentioned constraint. It is assumed that this value for ϵ_γ remains constant over the limited energy range (up to 2.5 MeV) above S_n considered here. The resulting values for $P_{p\gamma}$ are shown in Fig. 2. At low enough excitation energies, $P_{p\gamma}$ is seen to decrease as fewer γ-ray cascades emit a γ ray with energy greater than $E_{\rm th}$ and therefore ϵ_γ must decrease. An uncertainty of ±5–10% is assigned to the values of ϵ_γ used here. Unlike an approach based on discrete γ rays (see Ref. [20] for example), the fraction of cascades that pass through any particular transition need not be determined.

The exit-channel probability, $P_{p\gamma}$, determined using Eq. (5) for ^{156}Gd and ^{158}Gd using the different values of $E_{\rm th}$ considered here is shown in Fig. 2. The $P_{p\gamma}$ values are also compared to the results determined using γ rays from the $4^+ \rightarrow 2^+$ transition, which had the highest statistics of the discrete ground-state band transitions [20]. The results using the $4^+ \rightarrow 2^+$ transition and the statistical γ-ray approach are consistent in the energy region $S_n < E_{\rm ex} < S_n + E_{\rm th}$ for the different values of $E_{\rm th}$. The $P_{p\gamma}$ curves determined using the statistical γ-ray approach are seen to trend upward one by one at $E_{\rm ex} > S_n + E_{\rm th}$, as

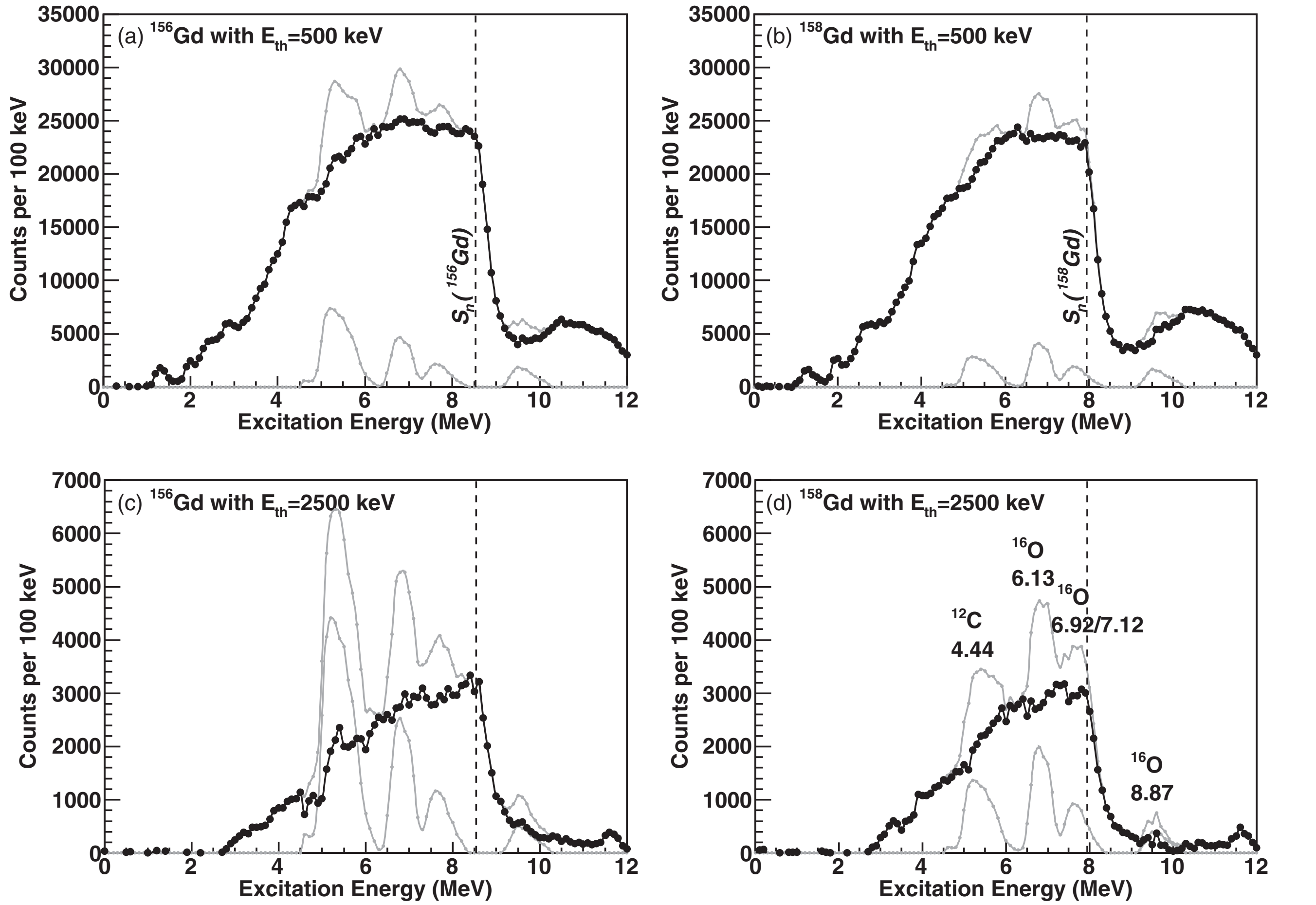

FIG. 1. The number of particle-γ coincidence events, $N_{p\gamma}$, using $E_{\rm th} = 500$ keV for (a) ^{156}Gd and (b) ^{158}Gd, and using $E_{\rm th} = 2500$ keV for (c) ^{156}Gd and (d) ^{158}Gd. In (d), the peaks due to inelastic scattering on ^{12}C and ^{16}O are labeled with the excited level energy in MeV. The peak structure (grey) is subtracted to determine $N_{p\gamma}$ due to the Gd isotopes (black). The neutron separation energies for ^{156}Gd and ^{158}Gd are indicated by the dashed lines.

background γ rays emitted following neutron emission can have energies greater than $E_{\rm th}$.

IV. COMPARISON WITH DISCRETE-TRANSITION DETECTION

A. Exit-channel detection efficiency

The Gd isotopes studied here are nearly ideal cases for measurements using discrete transitions because they have strong ground-state band collectors ($\approx$70% of the γ-ray cascades pass through the lowest 4^+ state) and the emitted γ rays have energies that can be efficiently detected using HPGe detectors. Even so, the statistical γ-ray approach used here has statistics that are up to a factor of 4 higher. For other nuclei that are more challenging to study by the discrete γ-ray analysis, the improvement could be much larger. Because the statistical γ-ray approach does not depend on energy resolution, an additional large increase in statistics could be obtained by using a different type of detector. Detector arrays that consist of scintillator detectors can more easily have nearly 4π solid angle coverage and a larger intrinsic detection efficiency and are well matched for the needs of this type of experiment. Using this type of detector array could increase the statistics collected by an additional order of magnitude.

B. Backgrounds

One challenge associated with using lower-resolution detectors or an analysis that makes only limited use of γ-ray energy information is that the approach is more susceptible to backgrounds from either contaminants or other reactions on the isotope of interest. Of course, many experimental properties (nuclear reaction, target purity, detector array and geometry, etc.) can have a significant effect on the backgrounds that are observed. In the analysis presented in this paper, only a limited energy window ($E_{\rm ex} < S_n + E_{\rm th}$) was free of backgrounds from (p, pn) reactions as can be seen in Fig. 2. At several values of $E_{\rm ex}$, large corrections for carbon and oxygen contamination also had to be made to both N_p and $N_{p\gamma}$. Additional backgrounds that could be particularly problematic would arise if decay channels other than γ-ray or neutron emission are open in the $E_{\rm ex}$ range being studied. For example, fission reactions would likely provide a significant γ-ray

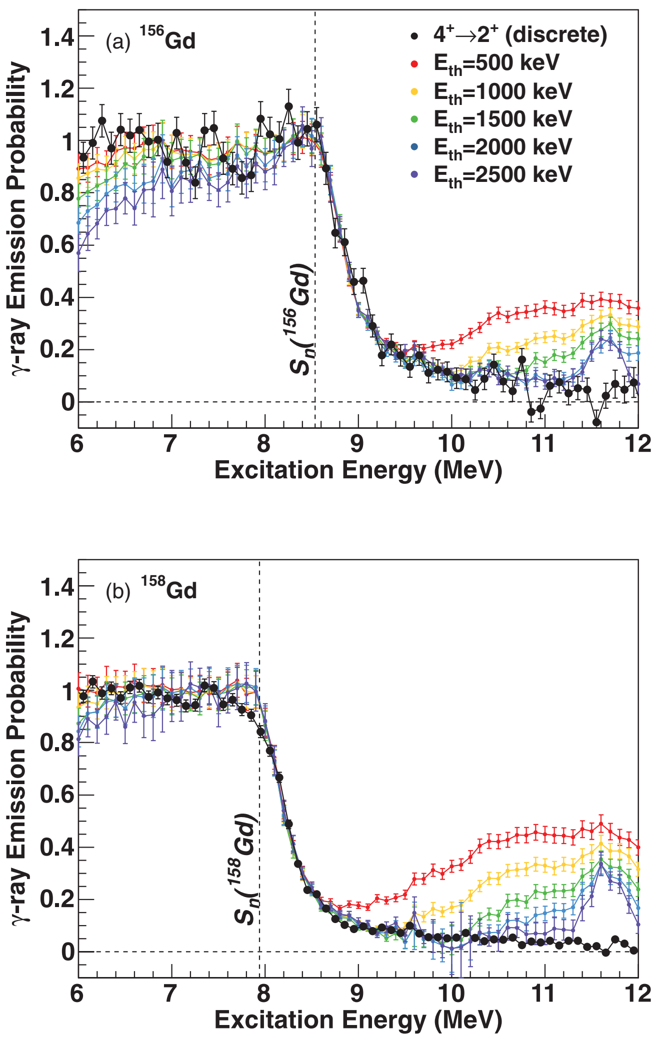

FIG. 2. (Color online) The exit-channel probability, $P_{p\gamma}$, for (a) ^{156}Gd and (b) ^{158}Gd compound nuclei for each E_{th} value. The parameter ϵ_γ is held constant at the values required to make $P_{p\gamma} = 1$ just below S_n for each curve.

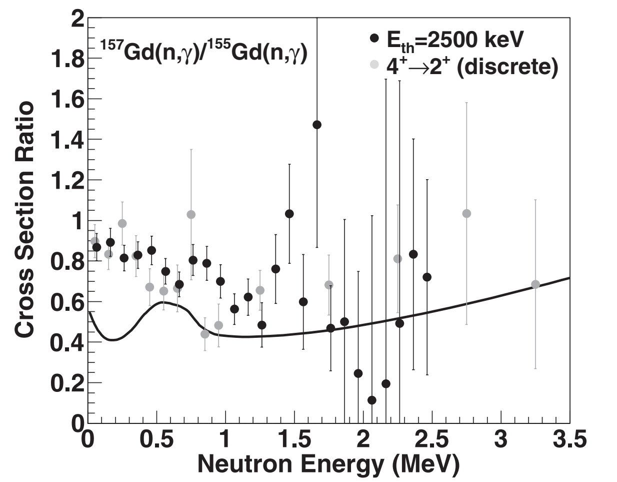

FIG. 3. The ratio of ^{157}Gd$(n,\gamma)/^{155}$Gd(n,γ) cross sections are shown for the surrogate analyses using statistical γ rays with $E_{\text{th}} =$ 2.5 MeV (black points) and discrete γ rays using the results for $4^+ \rightarrow 2^+$ transitions [20] (gray points). The smooth curve is the cross-section ratio that is calculated using reaction theory [20,25].

background that would make studies of actinide reactions difficult. With limited information on the energy of the emitted γ rays, any analysis must be performed with caution as it is more difficult to identify and quantify the experimental backgrounds.

C. Angular-momentum effects

In Ref. [20], Hauser-Feshbach-type calculations were carried out for the ^{155,157}Gd(n,γ) cross sections using relevant structure information (such as the energies of discrete levels, γ branching ratios, resonance information, etc.) obtained from the RIPL-3 database [37]. This work indicated that it was the angular-momentum mismatch that was responsible for the Weisskopf-Ewing approach yielding too large a result by factors of 2–3 for the ^{155,157}Gd(n,γ) cross sections when using $P_{p\gamma}$ determined from discrete γ-ray transitions.

The agreement between the values for $P_{p\gamma}$ obtained from discrete and statistical γ-ray approaches also reveals that the statistical approach suffers from the same angular-momentum effects discussed in detail in Ref. [20]. The consistency between the results of the two experimental approaches intuitively makes sense as both the low-lying discrete transitions and the statistical γ rays are emitted in the majority of decays and therefore observe nearly identical compound-nuclear J^π distributions. For example, at $E_{\text{ex}} > S_n$, the $4^+ \rightarrow 2^+$ transition is part of most decay cascades and therefore contains contributions from essentially all values of spin and parity.

The cross section ratio obtained for $E_{\text{th}} = 2.5$ MeV using Eq. (4) is shown in Fig. 3 and compared to results obtained from discrete γ rays. The results obtained for lower values of E_{th} are consistent with the 2.5-MeV result in the regions between S_n and $S_n + E_{\text{th}}$ that are free of contaminating reactions. The experimental results, however, are inconsistent with the cross-section ratio determined from the Hauser-Feshbach-type calculations that agree with direct measurement [20,25]. The ratio approach using the statistical γ-ray technique appears to fare no better than the discrete γ-ray technique—a factor of 2 difference between the direct measurement and the results of the surrogate approaches is observed up to corresponding neutron energies of several hundred keV. These results demonstrate that the results obtained from either discrete or statistical γ rays are susceptible to systematic shifts due to spin-parity mismatchs and the discrepancies do not, in general, cancel even when ratios of cross sections are considered.

Although the use of scintillator detectors could significantly increase the statistics collected in a surrogate nuclear reactions experiment, the limited energy resolution of these detectors results in less information obtained about the γ-ray cascade. The distribution of discrete γ-rays emitted in the γ-ray cascade provide a signature of the J^π distribution of the compound nucleus formed in the surrogate reaction. This information is very useful in interpreting the results of surrogate nuclear reaction experiments by providing important constraints needed to account for any J^π mismatch.

V. CONCLUSIONS

The surrogate nuclear reaction technique, which has been successfully applied to indirectly determine neutron-induced fission cross sections for actinides, is currently being applied to the study of radiative neutron-capture reactions. This approach uses measurements of the decay pattern of the excited compound nucleus of interest to determine (n,γ) cross sections. The determination of an (n,γ) cross section faces two significant challenges: (1) collecting the necessary γ-ray statistics at excitation energies above S_n and (2) accounting for the effects of any J^π mismatch between the desired capture reaction and the surrogate reaction used in the experiment.

An approach that seeks to identify the γ-ray exit channel from any energy deposition in the detector array greater than a selected threshold energy has been studied here. The γ rays that yield these signals are produced by complicated and varied γ-ray cascades that result from the de-excitation of highly excited nuclei and are referred to here as "statistical" γ rays. The results from a recent experiment that bombarded Gd targets with protons and detected the emitted γ rays with HPGe detectors were used in this analysis. The results obtained by identifying the γ-ray exit channel using statistical γ rays are found to be consistent with the results obtained using low-lying discrete γ rays. The statistical γ-ray approach, therefore, still suffers from the angular-momentum effects observed in other surrogate nuclear reaction experiments and corrections must be introduced to account for the differences. Also, the analysis must be performed with caution to ensure that all backgrounds (in particular from target contaminants, competing reactions, or fission in the case of actinide nuclei) are properly taken into account.

The statistical γ-ray approach, however, has the advantage that significantly higher statistics can be collected. Experiments can make use of lower-resolution detectors and therefore can be performed with detector arrays consisting of scintillators that can have higher efficiency and better solid angle coverage than typical HPGe detector arrays. This can be important for performing surrogate nuclear reaction experiments on nuclei that either have no strong collector transition or have a lot of highly converted transitions.

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