Simulating γ - γ coincidences of β -delayed γ -rays from fission product nuclei

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Abstract

Analyzing radiation from material that has undergone neutron induced fission is important for fields such as nuclear forensics, reactor physics, and nonproliferation monitoring. The γ -ray spectroscopy of fission products is a major part of the characterization of a material's fissile inventory and the energy of incident neutrons inducing fission.

Cumulative yields and γ -ray intensities from nuclear databases are inputs into a GEANT4 simulation to create expected γ -ray spectra from irradiated ^{235}U . The simulations include not only isotropically emitted γ -rays but also γ - γ cascades from certain fission products, emitted with their appropriate angular correlations. Here γ singles spectra as well as γ - γ coincidence spectra are simulated in detectors at both 90° and 180° pairings.

The ability of these GEANT4 Monte Carlo simulations to duplicate experimental data is explored in this work. These simulations demonstrate potential in exploiting angular correlations of γ - γ cascades in fission product decays to determine isotopic content. Analyzing experimental and simulated γ - γ coincidence spectra as opposed to singles spectra should improve the ability to identify fission product nuclei since such spectra are cleaner and contain more resolved peaks when compared to γ singles spectra.

Keywords: β -decay, γ spectroscopy, fission products, angular correlations, Monte Carlo

1. Introduction

The γ -ray spectroscopy of neutron-rich fission product nuclei is necessary for areas such as nuclear structure, nuclear forensics, reactor physics, nonproliferation monitoring, and dose rate predictions from reactor incidences. The identification of fission products is beneficial for determining fissile inventory and incident neutron energies. A measurement of key β -delayed γ -rays emitted from these isotopes is required, especially for fission nuclides in the “valley” and “wings” of the fission yield functions. However, given high activity and a large number of fission products, the γ -ray spectra often have too many peaks to resolve. Therefore, it is advisable to utilize spectra displaying only those γ -rays in coincidence with other specific γ -rays. For example, two γ -ray photons that arrive in a specific pair of germanium detectors within a μs time window. In such spectra the number of measured photons will be lower and so it becomes easier to resolve important photo-peaks. Thus it selects the decays desired and reduces unwanted background.

In this work we investigate pairs of γ -rays emitted with specific angular correlations in γ - γ cascades. We then look for the coincidences of γ -rays within detector

pairs at both 90° and 180° orientations. The purpose of these simulations is to investigate the possibility of exploiting angular correlations of γ - γ cascades to identify the type and the amount of specific isotopes present in a sample.

Here we compare the results from the simulations to experimental data from thermal neutron induced fission on a 93% enriched uranium foil. The irradiation occurred in a nuclear reactor with a thermal port, where the Cd ratio is $\approx 1/200$, at 90 kW of power for 200 minutes. A fraction of the irradiated and dissolved sample is then placed in a cylindrical vial of 1.5 cm in radius and the sample is about 5 mm in height.

2. GEANT4 simulations

A GEANT4 Monte Carlo simulation code [1] is used to simulate how γ -rays are emitted from and propagate through the cylindrical source and how they propagate through and deposit energy into adjacent detectors. The detector geometry consists of three almost identical high purity germanium crystals (HPGe, ORTEC GEM30P4-70) in a setup located in the radiochemistry complex at Lawrence Livermore National Laboratory.

There are two detectors 180° apart and a third detector at 90° to those two detectors. Thus there is one pair at 180° and two pairs at 90°.

The purpose of the simulations is to produce expected γ -ray spectra from decaying fission product nuclei in the sample described above. To that end, both the table of isotopes [2] as well as cumulative yield fission tables [3] are utilized to determine the amount and energy of photons to emit from the source. From the aforementioned databases isotopes were selected that have a cumulative yield above 0.4% and a half-life of one hour or greater. This range covers isotopes from ^{75}Ge up to ^{151}Pm , but the noble gases are excluded since many of those isotopes will escape before or during dissolution of the sample. For simplicity, once an isotope is selected it is assumed that the daughter and granddaughter isotopes are only populated through the decay of the parent until a stable isotope is reached. For example, if 1000 atoms of ^{140}Ba are populated then all 1000 of its decays will populate 1000 atoms of ^{140}La , which in turn decay to the valley of stability. Only Ba is the starting point in the mass A=140 chain because it is the first isotope with a half-life above one hour and a high enough cumulative yield as indicated above. All of the La is then populated only from Ba. In future simulations the populations of isotopes can include multiple starting points within a mass chain. For each isotope used at least three of the strongest γ -rays are included in the simulation. The half-lives and measurement time require the use of solutions to appropriate Bateman equations [4]. The total number of expected γ -rays emitted from each fission product is determined from the start and stop time for measuring and the cumulative yields. The simulations have a start time at 120 hours after irradiation ends and a completion time 12 hours later. This matches the experimental measurement times.

The GEANT4 code takes into account the photoelectric effect in detecting γ -ray photons, Compton scattering of the photons, and electron-positron production and annihilation. GEANT4 accesses databases with cross sections for these interactions based on the atomic number, density, etc. of the materials the photons pass through. At each step along which the photon (or other particle) is propagating, the simulation runs a Monte-Carlo process for each physics interaction that may or may not occur at that point. This is a standard way that physics interactions happen in Monte-Carlo codes like GEANT4 that both nuclear and high energy physicists utilize. In this project what we have changed is simply how the γ -ray photons are initiated.

Amongst these isotopes a number of them were

selected where both the isotope itself was produced abundantly in the fission [3] and a large amount of decay intensity passes through a pair of γ -rays in a γ - γ cascade. In the simulation these two γ -rays are emitted with a non-isotropic angular distribution between them. Based upon the multipolarity and spin-parity of the three levels involved in the cascade, an appropriate angular correlation function is chosen. Such a formula is based upon theoretical and experimental knowledge of the cascade [5].

A C++ code was written to create an input file to the simulation for those cascades. The energies of the two photons and the angular correlation function are inputs for this code. The photons are emitted with a gaussian distribution in energy with a sigma related appropriately to the energy of the photons. This is meant to mimic the appropriate energy resolution of the detectors in the simulation. A text file is formed, which is then used as input into the GEANT4 simulations. It tells the simulation to emit γ -ray one followed by γ -ray two, both emitted from the same point within the source volume. The two γ -ray photons are emitted with a specific angle between their initial directions. This process is repeated from random points within the cylindrical source and each pair is emitted with different angles between their vectors. The amount of pairs emitted at each angle follows the appropriate angular correlation function.

For example, in the decay of ^{60}Co , a common γ -ray calibration source, there are two γ -rays and each one is of multipolarity E2 with the following correlation function [5]:

$$W(\theta) = 1 + \frac{1}{8} \cos^2(\theta) + \frac{1}{24} \cos^4(\theta) \quad (1)$$

Here θ is the angle between the initial directions of the two γ -rays. This is one example where the spin and parity cascade is 4^+ to 2^+ to 0^+ . Certain even mass nuclei are included in this simulation because they are also present in the experimental data, including ^{132}I , that have this same angular correlation function for a prominent cascade of γ -rays in their decays. Some examples are seen in tables 1 and 2. There are also odd mass nuclei such as ^{99}Mo . It has a cascade of 739 keV followed by 181 keV. The spin and parities of the states involved are $1/2^+$ to $5/2^+$ to $9/2^+$. The angular correlation function for this cascade is the same as with ^{60}Co . However, in the decay of isotope ^{129}Sb there is a cascade of transition 683 keV followed by 544.7 keV, with spin and parities of $(9/2^+)$ to $5/2^+$ to $3/2^+$. It is a proposed E2 transition followed by an M1 and its angular correlation

function is as follows:

$$W(\theta) = 1 - \frac{3}{29} \cos^2(\theta) \quad (2)$$

If a γ - γ cascade has mixed multipolarity then more than one angular correlation function can be utilized based upon either a known mixing ratio between the multipolarities or evenly distributed amongst the two or three angular correlations required. For example, in the decay of ^{147}Nd there is the cascade of 319.4 keV followed by 91.1 keV. Both transitions are mixed M1+E2 and the spins and parities are $3/2^+$ to $5/2^+$ to $7/2^+$. To approximate the angular correlation function needed there are three functions used: one for an M1+M1 pair, another for an E2+E2 pair and a third for M1+E2. However, to improve upon this one can take into account the fact that the measured mixing ratio for the M1/E2 strength is 0.41(3) for the 319.4 keV transition and 0.082(10) for the 91.1 keV transition [6]. The cascade used in ^{97}Zr decay is in a similar situation, as shown in Table 1.

2.1. Photo-peak efficiencies in simulation vs. experimental data

Before running the simulation, the detector geometry was designed to match specifications given by the manufacturer of the HPGe detectors. The size, position and volume of the germanium matches the manufacturer's details as well as our placement in the real experimental setup. Aluminum housings, cryostat inserts, the beryllium window, etc. are made of the correct material and density in the simulation to match the experimental setup. However, slight modifications had to be made in order to match the experimental efficiency curve with the same curve in simulation. The main alteration was to make aluminum housings around the germanium thicker than stated by 1-3 mm. Such alterations are commonly needed in simulations as detector specifications, such as dead layers and crystal size, can be incorrect in the manufacturer's manuals. This has also been addressed in other simulation work in literature [7]. Figure 1 displays the efficiency of one HPGe detector within the simulation, where a standard γ calibration point source containing the radionuclides ^{241}Am , ^{109}Cd , ^{57}Co , ^{133}Ba , ^{137}Cs , ^{60}Co , ^{88}Y , and $^{152,154}\text{Eu}$ was placed one centimeter from the front face of the upward facing detector. This efficiency simulation matches the conditions of the efficiency measurements. More specifically, the plastic point source dimensions and material were duplicated in simulation to match experiment. Any self absorption that would occur in experimental efficiency measurements is taken into account in the propagation

of γ -rays through the plastic source in simulation.

An experimental, absolute efficiency curve was obtained from a series of efficiency measurements that take into account any summing of γ -rays at close distances and removing this effect from the final efficiency curve. In the efficiency simulation the summation of γ - γ cascades is not an issue because individual γ -rays with desired energies are emitted in the simulation one at a time, whereas in a real measurement it is impossible to separate γ -rays from their cascade partners and hence they can appear summed in a single event.

Figure 1 shows experimental efficiency points (with error bars) from this curve but at the same energies that were input energies into the simulated efficiency measurement. Errors from the simulation are slightly less than 1%. A comparison with the measured efficiency is shown in Figure 2. This figure displays the residuals, or percent difference, between efficiency in the simulation and efficiency in the experiment as a function of γ energy. The two efficiencies are in good agreement, except it was difficult to match the low energy peak in simulation versus experiment. This is off by 10-12%, but taken into account in the analysis discussion.

3. Simulation results and comparison to experimental data

The purpose of these simulations is to produce coincident γ -ray spectra likely to be measured from a source of ^{235}U irradiated with thermal neutrons, as was done to produce the experimental data. The simulation emits and propagates γ -rays that are emitted after the β -decays of fission product nuclei. The total number of fissioning parent ^{235}U nuclei used to create numbers of fission product nuclei to place into simulation is 1×10^{10} . This number is chosen to obtain a sufficient amount of statistics in simulation such that peaks of interest appear abundantly enough for a comparison to experimental data. Matching an absolute number of fission products in simulation to experiment is not important because in this work the typical comparisons are made with ratios of counts in a γ -ray peak to another peak from one isotope in particular, typically ^{99}Mo but others can be utilized.

Table 2 contains those fission products where γ - γ cascades are included. It indicates each pair of γ -rays used for non-isotropic emission in the simulation. The coincidence condition in simulation is met when a γ -ray is seen after another one in the detector of interest. For example, the 739 keV transition in ^{99}Mo decay is followed by the 181 keV transition in cascade. If the 739 keV γ is observed as an event after the detection of the 181 keV

Isotope	γ cascade (keV)	multipolarities/spin-parities	angular corr. func.
^{97}Zr	1147.97, 602.37	M1,E2+M1,E2 ($5/2^+$ to $7/2^+$ to $9/2^+$)	$1 + \frac{1}{13} \cos^2(\theta); 1 + 0.19 \cos^2(\theta); 1 - 0.54 \cos^2(\theta) + 0.46 \cos^4(\theta)$
^{99}Mo	739, 181.0	E2+E2 (4^+ to 2^+ to 0^+)	$1 + \frac{1}{8} \cos^2(\theta) + \frac{1}{24} \cos^4(\theta)$
^{132}I	667.7, 772.6	E2+E2 (4^+ to 2^+ to 0^+)	$1 + \frac{1}{8} \cos^2(\theta) + \frac{1}{24} \cos^4(\theta)$
^{132}Te	111.76, 116.3	M1,E2+M1,E2 (1^+ to 2^+ to 3^+)	$1 + \frac{1}{13} \cos^2(\theta)$
^{133}I	856.0, 267.0	E2+E2 ($9/2^+$ to $5/2^+$ to $1/2^+$)	$1 + \frac{1}{8} \cos^2(\theta) + \frac{1}{24} \cos^4(\theta)$
^{140}La	1596.21, 487.021	E2+E2 (4^+ to 2^+ to 0^+)	$1 + \frac{1}{8} \cos^2(\theta) + \frac{1}{24} \cos^4(\theta)$
^{140}Ba	304.8, 162.6	M1,E2+M1,E2 (1^- to 2^- to 3^-)	$1 + \frac{1}{13} \cos^2(\theta)$
^{143}Ce	350.6, 139.0	E2+E2 ($7/2^+$ to $3/2^+$ to $7/2^+$)	$1 + \frac{30}{582} \cos^2(\theta)$
^{147}Nd	91.1, 319.4	M1,E2+M1,E2 ($3/2^+$ to $5/2^+$ to $7/2^+$)	$1 + \frac{1}{13} \cos^2(\theta); 1 - 0.38 \cos^2(\theta) + 1.02 \cos^4(\theta); 1 + 0.19 \cos^2(\theta)$

Table 1: A list of the multiplicities, spin-parities, and angular correlation functions for various cascades, some of which are also shown in Table 2. For the isotopes ^{97}Zr and ^{147}Nd it is shown that one can utilize more than one function to create the angular correlations, as explained in the text.

Figure 1: The experimental and simulated photo-peak efficiency curves from a calibration source placed one centimeter from the front of the upward pointing HPGe detector. The error bars are about 1% in size for experimental values and smaller for simulation values.

Figure 2: The ratios of simulated over experimental photo-peak efficiencies. The majority of the energy range shows agreement within 10% of experimental values, as can be seen with 'guidelines' at $\pm 10\%$. Error bars are included.

γ in another detector then a count is placed in a two dimensional γ - γ coincidence matrix at coordinates 181 keV on the x-axis and 739 keV on the y-axis. Another count is placed at coordinates 181 keV on the y-axis and 739 keV on the x-axis to make it symmetric. Thus when an x-projection or y-projection is created from the matrix at 739 keV, a one dimensional γ spectrum is produced and reveals a peak at 181 keV. Likewise, a projection on the 181 keV transition produces a one dimensional spectrum with a peak at 739 keV. Thus a standard coincidence matrix is made in simulation, just as is standard with γ - γ coincidence measurements.

In Table 2 there are γ -ray transitions that appear in a full range, one-dimensional projection of the whole γ - γ coincidence matrix. The ratio of counts in the simulation matrix over counts in the experimental matrix are shown. These ratios are modified if the simulated efficiency of detection is over 10% different than the experimental value. For example, the two transitions in ^{132}Te decay are 111.76 and 116.3 keV, whose simulated efficiencies of detection are about 10% higher than the measured values with the calibration source. The same applies to 91 keV from ^{147}Nd decay, whose efficiency of detection is overestimated in simulation by 12%.

There is a wide range of values for these ratios. There are a number of caveats to consider when attempting to simulate the emission and propagation of these γ -rays. In some cases there may be γ with γ or γ with x-ray summing that can alter the ratio of simulated over ex-

perimental counts since it is difficult to reproduce this in simulation while maintaining non-isotropic angular correlations. Both ^{132}Te and ^{99}Mo appear to be overestimated in simulation, while isotopes such as ^{140}La are not. Another issue is that some regions are overcrowded with difficult to resolve γ -ray transitions, even in the coincidence spectra. Such is the case in the 700-800 keV region. This may explain why in ^{132}I decay the 667.7 keV transition is well duplicated in simulation but the 772.6 keV transition is slightly over estimated even though they are in cascade with each other. Furthermore, because of chemical dissolution of the sample before measuring the radiation, volatile elements such as Te and I may be lost and hence the quantity will be altered from what is given by fission yield tables [3]. Also, some γ -ray transitions from isotopes abundantly produced in fission are intense enough to appear as accidental coincidences. They will appear in coincidence with γ -rays that are not in a cascade with them. One example is the strong transition of 1596 keV in the decay of ^{140}La . Finally, the 350.6 keV peak in ^{143}Ce decay overlaps a common room background γ -ray peak at 351.9 keV.

The average ratio of simulated over experimental counts found in these coincidences is only 1.05, although the standard deviation is 0.46. Even though there is a wide spread in these ratios there are a number of caveats explained above. The most important point is how these numbers may change when fission occurs in

Figure 3: The experimental low energy region of the x-projection on the γ - γ coincidence matrix. The 111.76 and 116.3 keV γ peaks from ^{132}Te decay are visible as one example.

Figure 4: The simulated low energy region of the x-projection on the γ - γ coincidence matrix. The γ -rays 111 and 116 keV in ^{132}Te decay are visible in the simulation. The 181 keV peak from ^{99}Mo and the 228 keV peak from ^{132}Te are also present.

Isotope	γ -ray (keV)	ratio sim./exp. (error)	ratios normalized to ^{132}I	ratios normalized to ^{99}Mo
^{99}Mo	181.0	1.45 (0.02)	1.32 (0.02)	1.12 (0.02)
^{99}Mo	739.0	1.29 (0.02)	1.18 (0.01)	1.00 (0.01)
^{132}I	667.7	1.090 (0.003)	1.000 (0.003)	0.850 (0.002)
^{132}I	772.6	1.330 (0.004)	1.212 (0.004)	1.020 (0.003)
^{132}Te	111.76	1.76 (0.10)	1.65 (0.09)	1.39 (0.08)
^{132}Te	116.3	1.48 (0.08)	1.35 (0.07)	1.14 (0.06)
^{140}La	1596.21	0.897 (0.004)	0.817 (0.004)	0.690 (0.003)
^{140}La	487.021	1.09 (0.01)	1.001 (0.005)	0.850 (0.004)
^{140}Ba	304.8	1.25 (0.03)	1.14 (0.03)	0.96 (0.02)
^{140}Ba	162.6	0.94 (0.02)	0.86 (0.02)	0.73 (0.01)
^{143}Ce	350.6	0.61 (0.05)	0.56 (0.05)	0.47 (0.04)
^{143}Ce	139.0	0.314 (0.001)	0.286 (0.001)	0.240 (0.001)
^{147}Nd	319.4	0.83 (0.04)	0.76 (0.03)	0.64 (0.03)
^{147}Nd	91.1	0.33 (0.01)	0.34 (0.01)	0.250 (0.005)
Average ratios		1.05 (0.02)	0.98 (0.02)	0.83 (0.02)

Table 2: A list of fission product nuclei and their prominent γ -rays in the simulation of ^{235}U thermal fission product γ spectroscopy. These are counts as seen in a full range, one-dimensional energy projection of the γ - γ coincidence matrix. The first column shows the isotope in question and the second column indicates the γ -ray energy. The third column shows the ratio of counts in the simulation over the counts in that same peak in the experimental data. In this column the ratios for both transitions in ^{132}Te and the 91 keV transition in ^{147}Nd decay are modified for the known efficiency differences between simulation and experiment. Column four shows the same ratios but normalized to the peak at 667.7 keV in ^{132}I since it was the closest in the absolute ratios. Column five shows the ratios normalized to the peak at 739.0 keV in ^{99}Mo , a typical isotope for determining total fissions. The uncertainties on the last digits of the ratios are in parentheses.

Figure 5: The energy region from 500 to 1000 keV showing many γ -ray lines in simulation. This figure shows the result from simulations that include angular correlations of emitted γ -rays.

Figure 6: The same energy region from simulation as shown in figure 5 but with all photons emitted isotropically.

Figure 7: The experimental, full energy range of the x-projection on the γ - γ coincidence matrix. The higher energy regions can be compared to simulations in figures 5 and 6.

Figure 8: The simulated, full energy range of the x-projection on the γ - γ coincidence matrix. The Compton background is the main discrepancy it has with the full experimental projection. This is because of both a difference in the absolute number of fission products in simulation vs. experiment and because not all of the higher energy, lower intensity peaks from fission products are included. High energy background lines are also not included.

a different fuel or with a different neutron energy distribution. It is relevant to include angular correlations in the simulation of fission product γ -ray spectroscopy. Such angular correlations should not always be treated as simply isotropic, as some of these functions for decays in ^{99}Mo , ^{132}Te , ^{132}I , and ^{140}La are not “shallow” as a function of angle.

To add to this discussion there are also shown ratios of counts in these peaks in simulation versus experiment. However, these ratios are normalized to specific isotopes. In table 2 there is a column showing those ratios normalized to the peak at 667.7 keV in ^{132}I . This isotope is chosen because its peak has the closest agreement with the intensity of all peaks that have an angular correlation function included here. The average ratio value with this normalization is 0.98. Here the transition of 739 keV in ^{99}Mo decay is utilized as another example. The average ratio for that normalization is 0.83. The average ratio of simulated over experimental fission γ -ray peaks for this overall analysis is then 0.95 and the average standard deviation for all three sets of ratios is 0.43.

4. Symmetric γ - γ matrix test

In these simulations we have operated under the assumption that the γ - γ matrices are symmetric. For two γ -rays that are in cascade, call them A and B, the γ -ray A should be detected the same amount in relation to γ -ray B regardless of which γ -ray is detected first. In an experiment this is of course true because with the slow rise time of HPGc detectors and picosecond or femtosecond lifetimes of non-isomeric nuclear states the two γ -rays appear to arrive “simultaneously” in the detectors.

Furthermore, in our setup the detectors were almost identical (same material, size, efficiency, etc.) and the two detectors that are 180° apart are the same distance from the source. Since the distance from the source to both back-to-back detectors is the same and the distances from one side detector to the bottom detector, 90° pair, is the same as for the other 90° pair, any γ - γ matrix made from coincident pairs in any detector pairing should be symmetric.

However, this assumption may not be true in simulations. To test this a single series of γ - γ emissions representing cascades from a decay are simulated again. First, there is a condition in the code requiring γ -ray A

Figure 9: The one dimensional spectrum from a non-symmetric γ - γ matrix. Here the 181 keV γ -ray is gated on as the first transition in the cascade from ^{99}Mo decay. The 739 keV transition is the cascade partner.

Figure 10: The one dimensional spectrum from a non-symmetric γ - γ matrix. Here the 739 keV γ -ray is gated on as the first transition in the cascade from ^{99}Mo decay. Within error bars, as explained in the text, photo-peaks in this spectrum are identical to the ones in Figure 9.

is detected first and then any γ -ray B (any energy deposition greater than zero) is detected in another detector. Their energies are recorded in a matrix but this matrix is non-symmetric. That is, a point is placed at coordinates A (energy of γ -ray A) on the horizontal axis and B (energy of γ -ray B) on the vertical axis. However, no point is placed at the alternative coordinates of B on the horizontal axis and A on the vertical. A one dimensional spectrum is produced from a full projection onto the horizontal axis.

This exercise is repeated but with a condition requiring γ -ray B is detected first and then any γ -ray energy deposited afterwards is recorded as γ -ray A in a non-symmetric matrix in the same manner as before. Another one dimensional spectrum is produced from the full projection onto the horizontal axis of this second γ - γ matrix. Now there exist two γ spectra (one dimensional spectra). The first shows all γ -rays in coincidence with γ -ray A and the second shows all γ -rays in coincidence with γ -ray B. An example of this was run with the cascade in ^{99}Mo decay (181 keV and 739 keV) and is shown in Figures 9 and 10.

Figure 9 shows the spectrum acquired from requiring the 181 keV peak to be the first in cascade detected in a detector. Here the counts in the 181 keV photo-peak are 578.5(24.6) and the counts in the 739 keV peak are 439.0(21.2).

Figure 10 shows the spectrum acquired from requiring 739 keV to be detected first. There the counts in the 181 keV peak are 581.4(24.6) and in 739 keV the counts are 456.4(21.4). Hence within error bars the counts in the 181 keV peak, regardless of which transition is detected first, are the same. The same is true of the 739 keV peak. Finally, we have verified such numbers for both the 180° pair as well as the 90° pairs of detectors. Both angular combinations do produce symmetric matrices in this work. The same procedure was performed on the two transitions in a prominent cascade in ^{140}La . The 487 keV and 1596 keV transitions are the same, within error bars, regardless of which of the two transitions were detected first in simulation.

Therefore the γ - γ matrix created in simulation is symmetric, but one should always double check with similar simulation techniques as described above to be certain that the efficiency of detecting γ -ray one followed by γ -ray two is the same as γ -ray two followed by γ -ray one.

5. Conclusion

This paper has covered the potential of exploiting angular correlations of γ - γ cascades in coincidence data

to help identify isotopes present in an unknown sample for nuclear forensics and other purposes. The average ratio of counts in simulation vs. experiment ranges from 0.83 to 1.05 for the three scenarios considered, with an average of those values at 0.95. There is still a large statistical spread in those ratios, but this is due to other problems one must overcome in future data analysis such as photon summing, background γ -ray transitions, chemical alterations, accidental coincidences, etc. This paper has also addressed the assumption of using symmetric γ - γ matrices for coincidences in simulation. It appears to be a safe assumption, but one can always verify this with simple, supplementary simulations as described above.

Furthermore, we have utilized thermal neutrons inducing fission in ^{235}U because γ -ray intensity ratios for this specific neutron spectrum are best understood in nuclear forensics applications as opposed to fission and high energy neutrons inducing fission. Thus it is an excellent starting place to explore angular correlations of γ -rays from fission product nuclei. However, in the future we will need to design experiments where fission neutrons and high energy neutrons are utilized for producing the same nuclei but with different fission yields. It is more experimentally challenging, but work is ongoing at facilities such as the Device Assembly Facility in Nevada and various other reactors.

Isotopes that have an angular correlation such as is seen in the E2-E2 γ - γ cascade in ^{60}Co decay are excellent candidates for exploiting coincidence data in identifying fission product nuclei of unknown samples. This is because that particular correlation function is not as shallow as many other functions and the coincidences should be more pronounced in 180° vs. 90° detector pairs. Also, the symmetric nature of the γ - γ matrices has been verified within this work. Future simulations will focus more on isotopes in the ^{103}Ru region, as this mass region has a significant difference in fission yields between ^{235}U and ^{239}Pu thermal fission over other mass regions. Isotopes in the “wings” of the fission yield plots will also be explored. Such mass regions as the “valley” and “wings” vary more with different fuels and different incident neutron energies. Hence exploitation of angular correlations in isotopes such as ^{91}Sr , ^{92}Y , $^{103,105}\text{Ru}$, ^{112}Ag , ^{153}Sm , etc. will be explored in the future with more computational power and more experiments but with the same technique outlined in this paper.

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