

Effect of pre-equilibrium spin distribution on neutron induced ^{150}Sm cross sections

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Abstract. Prompt γ -ray production cross section measurements were made as a function of incident neutron energy ($E_n = 1$ to 35 MeV) on an enriched (95.6%) ^{150}Sm sample. Energetic neutrons were delivered by the Los Alamos National Laboratory spallation neutron source located at the Los Alamos Neutron Science Center (LANSCE) facility. The prompt-reaction γ rays were detected with the large-scale Compton-suppressed Germanium Array for Neutron Induced Excitations (GEANIE). Above $E_n \approx 8$ MeV the pre-equilibrium reaction process dominates the inelastic reaction. The spin distribution transferred in pre-equilibrium neutron-induced reactions was calculated using the quantum mechanical theory of Feshbach, Kerman, and Koonin (FKK). These pre-equilibrium spin distributions were incorporated into the Hauser-Feshbach statistical reaction code GNASH and the γ -ray production cross sections were calculated and compared with experimental data. Neutron inelastic scattering populates ^{150}Sm excited states either by (1) forming the compound nucleus $^{151}\text{Sm}^*$ and decaying by neutron emission, or (2) by the incoming neutron transferring energy to create a particle-hole pair, and thus initiating the pre-equilibrium process. These two processes produce rather different spin distributions: the momentum transfer via the pre-equilibrium process tends to be smaller than in the compound reaction. This difference in the spin population has a significant impact on the γ -ray de-excitation cascade and therefore in the partial γ -ray cross sections. The difference in the calculated partial γ -ray cross sections using spin distributions with and without pre-equilibrium effects was significant, e.g., for the 558-keV transition between 8^+ and 6^+ states the calculated partial γ -ray production cross sections changed by 70% at $E_n = 20$ MeV with inclusion of the spin distribution of pre-equilibrium process.

1 Introduction

Recent results of γ -ray production cross section measurements at LANSCE performed with the GEANIE detector array [1] demonstrated that the spin distribution of the pre-equilibrium reaction has a large impact on the γ -ray transition probability when incident energies are above $E_n = 10$ MeV [2,3]. Since it was previously assumed that the spin distribution in the pre-equilibrium process had a limited impact on nuclear reaction cross sections, classical theories such as the exciton model [4,5] are still widely used to analyze particle emission data at high incident energies. A realistic treatment of the spin distribution improved the accuracy of calculations of γ -ray production cross sections with the statistical Hauser-Feshbach model.

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a significant impact on the de-excitation γ -ray cascade. The spin-distribution in the pre-equilibrium process is calculated with the FKK model, and the calculated spin-distribution is combined with the GNASH Hauser-Feshbach statistical model calculations [6]. To examine the influence of the FKK calculation, we also consider the case with the spin-distribution in the pre-equilibrium process the same as for the compound process. In the past such an assumption has often been made for Hauser-Feshbach plus exciton model calculations. We present comparisons of γ -ray production cross sections for neutron-induced reactions on ^{150}Sm .

2 Experimental setup and data analysis

The experimental data were obtained at the LANSCE Weapons Neutron Research (WNR) facility. At the WNR facility, spallation neutrons are produced by bombarding a natural W target with an 800-MeV pulsed proton beam from the LANSCE linac. The pulsed proton beam consists of micropulses 1.8 μs and 3.6 μs apart, bunched into macropulses 625 μs in duration. Spallation neutrons with energies ranging from a few keV to nearly 800 MeV are produced. The scattering sample consisted of 7 mg of Sm_2O_3 in the form of disks

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2.4 cm diameter, enriched to 95.6% in ^{150}Sm . The γ -rays were detected with the GEANIE spectrometer, located about 20 m from the neutron source on the 60° right flight path. For this experiment, the GEANIE spectrometer consisted of 11 planar and 15 25% High-purity Ge (HPGe) coaxial detectors. All of the planars and 9 of the coaxial detectors were equipped with Compton suppression shields. The planar detectors were used to measure γ -rays with energies less than 1 MeV and the coaxial detectors to measure γ -rays with energies up to 3 MeV. The efficiency of the array has been calibrated through a series of source measurements, supplemented by detailed modeling [7] using the transport code MCNP [8]. The neutron flux was determined using a fission chamber consisted of $^{235,238}\text{U}$ foils [9] located 2 m upstream from the GEANIE spectrometer. Neutron energies were determined by the time-of-flight (TOF) technique, using the detection time of the “flash” of γ rays caused by the spallation reaction with respect to the beam rf signal as a reference marker. The excitation functions were obtained by applying TOF gates 15 ns wide on the γ -ray events in the interval to $E_n = 1$ to 35 MeV. For each TOF bin a 1D γ -ray pulse-height spectrum was generated.

The data were collected with two different (1.8 and 3.6 μs) spacing between beam pulses. Due to a “wrap-around” problem (the flight path is long enough that low energy neutrons arrive at the target location at the same time as high-energy neutrons from a successive pulse) the 334-keV transition between 2^+ and 0^+ in ^{150}Sm was not extracted from the 1.8 μs spacing runs. In order to resolve this problem a one-day run with 3.6 μs spacing was performed. The partial γ -ray cross sections of $^{150}\text{Sm}(n, n'\gamma)^{150}\text{Sm}$ for $E_n = 1 - 35$ MeV extracted from 3.6 μs spacing data were compared with the 1.8 μs spacing data [10]. The comparisons of the prominent γ -ray cross sections, for example 439-keV ($4^+ \rightarrow 2^+$) and 505-keV ($6^+ \rightarrow 4^+$) γ rays, are extracted from both sets of data and cross sections from two data sets agreed very well.

To confirm our experimental and analysis techniques, the partial cross section of the $2_1^+ \rightarrow 0_1^+$ transition in ^{56}Fe was extracted from a series of runs with the ^{150}Sm sample sandwiched between 5-mil ^{nat}Fe foils. This partial cross section has been extracted from both planar and coaxial data, and is reported in ref. [10]. These data are compared to the cross section of 705 ± 56 mb at $E_n = 14.5$ MeV, evaluated by Nelson et al. [11]. The partial cross sections for 847-keV were consistent [$\sigma_\gamma(847) = 719 \pm 11$ mb for planar detectors at $E_n = 14.5$ MeV], within errors, with evaluated work.

3 Theoretical calculations

The γ -ray production cross sections were calculated with the GNASH code [6]. GNASH calculates the pre-equilibrium process with the exciton model, which is based on a classical theory and does not calculate spin transfer. However, it is known that the exciton model gives the fraction of pre-equilibrium to total particle emission reasonably well. We employ the exciton model for the pre-equilibrium strength calculation, but then modify the spin-distribution as calculated with the FKK theory. The MSD calculation employed is similar to the modeling of Koning and Chadwick [12], and is reported elsewhere [13]. In the present analysis, the

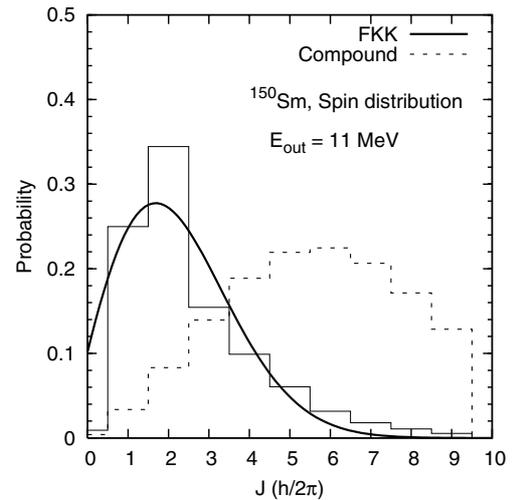


Fig. 1. Comparison of the spin-distribution for $E_{in} = 20$ MeV and $E_{out} = 11$ MeV, calculated with the FKK model (solid histogram), and the compound reaction (dotted histogram). The smooth curve is a Gaussian fit to the FKK result.

MSC component is assumed to have the same spin distribution as the compound process, because it has a weak dependence on the angular distribution, and the magnitude is usually smaller than the MSD [14] contribution.

The one-step calculation of MSD gives the spin-dependent population of continuum states in ^{150}Sm . Since the ground state spin of ^{150}Sm is zero, the spin distribution in the continuum populated by the one-step process is the same as the J -dependence of the MSD angle-integrated cross sections. We also calculate the population by a pure compound reaction. The initial population of ^{150}Sm (after neutron inelastic scattering, but before γ -ray cascading) is a sum of pre-equilibrium and compound contributions.

The calculated one-step FKK spin-distributions are expressed by a Gaussian form

$$R_{\text{MSD}}(J) = \frac{J + 1/2}{\sigma^2} \exp \left\{ -\frac{(J + 1/2)^2}{2\sigma^2} \right\}, \quad (1)$$

where σ^2 is a spin cut-off parameter. An example is shown in figure 1, which shows the calculated spin-distribution for ^{150}Sm inelastic scattering at a neutron incident energy of 20 MeV and an emitted neutron energy of 11 MeV. The solid histogram is the FKK result, and the dotted histogram is the spin-distribution of the compound reaction. The FKK spin-distribution is peaked at lower J -values – a high-spin state is difficult to make with a simple $1p-1h$ configuration in a single-particle model.

We fit the Gaussian form of equation (1) to the FKK results with various neutron incident/outgoing energies to obtain σ^2 as a function excitation energy E_x . Figure 2 shows the spin-distributions in the continuum of ^{150}Sm that is excited by 20-MeV neutron inelastic scattering. In past nuclear model calculations, the spin-distribution in the pre-equilibrium process has been assumed to be the same as in the compound reaction. This is illustrated in the top panel of figure 2. With the quantum mechanical theories of the pre-equilibrium process, the spin-distribution can be calculated on a more

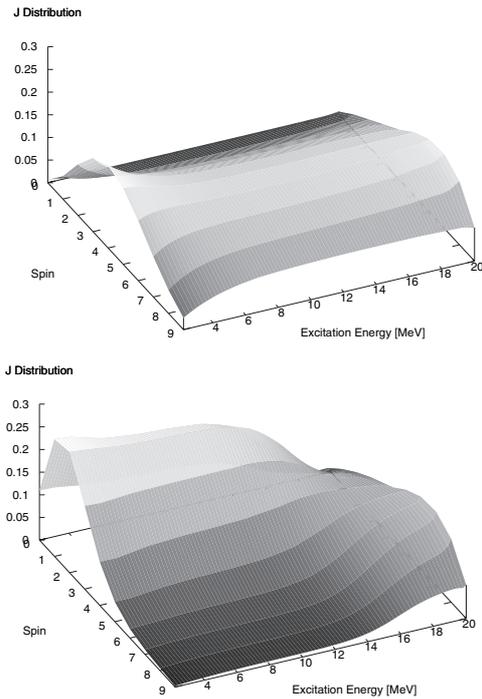


Fig. 2. The spin distributions in excited ^{150}Sm after neutron inelastic scattering, with different assumptions for the pre-equilibrium spin transfer. The neutron incident energy is 20 MeV. The upper panel shows the case when the pre-equilibrium spin distribution is assumed to be the same as the compound reaction. The bottom panel illustrates the case when the FKK spin distribution is included in the GNASH calculations.

realistic basis. This is shown in the bottom panel. The excited nucleus has a spin-distribution that is peaked at lower J -values when the excitation energy of the residual state is not too high. The statistical Hauser-Feshbach model code, GNASH was used to calculate the γ -ray production cross sections for neutron inelastic scattering on ^{150}Sm . The particle transmission coefficients were calculated using the coupled channel optical potentials of Kuneida et al. [15] for neutrons and protons, and the α -particle optical potential of Avrigeanu et al. [16] was adopted for the α particles. The level scheme of ^{150}Sm and the γ -ray branching ratios were taken from Table of Isotopes [17] and RIPL-2 [18]. We included the discrete levels of ^{150}Sm up to 1.836 MeV (8^+) in the calculation.

4 Results and discussions

Comparisons of the partial γ -ray cross sections for the 439 keV (4^+ to 2^+), 505 keV (6^+ to 4^+), and 558 keV (8^+ to 6^+) ground state band transitions in ^{150}Sm are shown in figures 3–5. For higher-spin residual states, for example in figures 3–4, the inclusion of the FKK spin distribution has a large effect on the γ -ray production cross sections, reducing the calculated production cross sections above $E_n = 10$ MeV. The FKK calculation has lower angular momentum transfer to the residual nucleus, and the population of the high-spin states in the continuum is reduced. Therefore, the cross sections for the γ -ray transitions from high-spin states are strongly

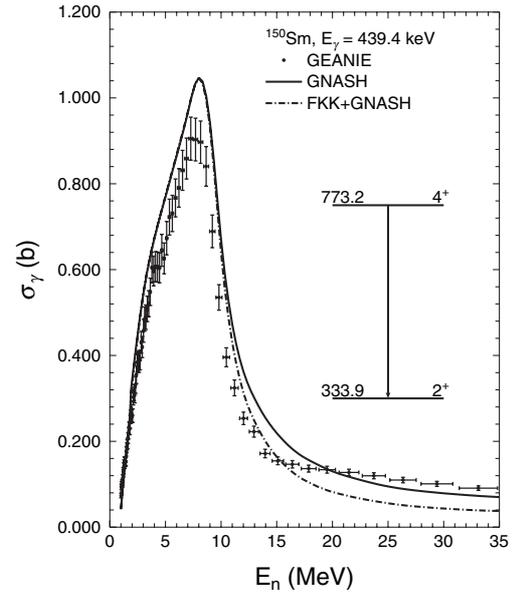


Fig. 3. Comparison of the 439-keV γ -ray production cross section with calculation. The solid line represents the FKK + GNASH calculation, and the dashed line represents the case where the pre-equilibrium spin-distribution is assumed to be the same as for the compound reaction.

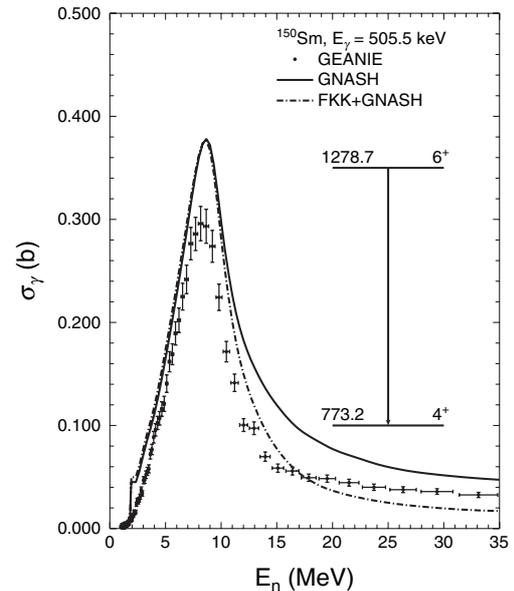


Fig. 4. Same as figure 3 but for the 505-keV γ -ray.

suppressed. Larger effect is seen in the higher-excited 8^+ states, as example of which ($8^+ \rightarrow 6^+$) is shown in figure 5. The difference in the cross sections between including spin distributions with and without pre-equilibrium effects is significant. The 558-keV transition between 8^+ and 6^+ states the calculated partial γ -ray production cross sections changed by 70% at $E_n = 20$ MeV with inclusion of the spin distribution of pre-equilibrium process which consistent with the measured experimental data.

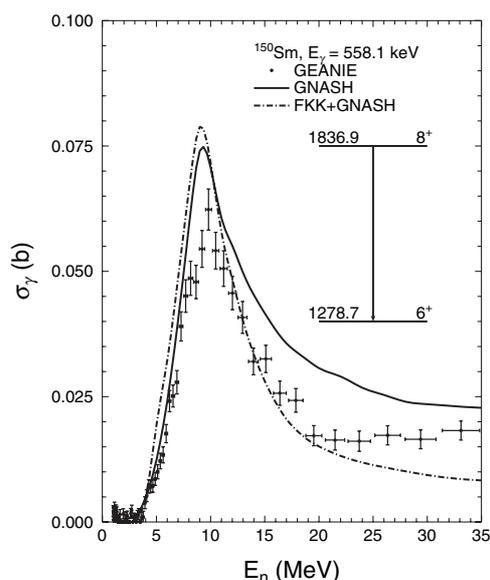


Fig. 5. Same as figure 3 but for the 558-keV γ -ray.

5 Conclusion

Excitation functions of prompt γ -rays produced in the $n + {}^{150}\text{Sm}$ reaction have been measured using the GEANIE spectrometer at the LANSCE/WNR facility. Partial γ -ray cross sections for $n + {}^{150}\text{Sm}$ reactions were calculated using the Hauser-Feshbach code GNASH and GNASH-FKK for neutron energies up to 35 MeV. The calculation includes the pre-equilibrium processes for the first emitted particle and produces activation cross sections, population cross sections for isomeric states and production cross sections for γ -rays from low-lying discrete excited states. The spin distribution of the pre-equilibrium process in ${}^{150}\text{Sm} + n$ reactions was calculated using the quantum mechanical theory of FKK. The FKK spin distribution was incorporated into GNASH calculations and the γ -ray production cross sections were calculated and compared with experimental data. The probability of γ -ray transitions from a high spin state is strongly suppressed because of the pre-equilibrium spin distribution which agree with measured experimental data.

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References

1. J.A. Becker, R.O. Nelson, Nucl. Phys. News Int. **7**, 11 (1997).
2. T. Kawano, P. Talou, M.B. Chadwick, Nucl. Instrum. Meth. A **562**, 774 (2006).
3. D. Dashdorj et al. (in press Phys. Rev. C) (2007).
4. J.J. Griffin, Phys. Rev. Lett. **17**, 478 (1966).
5. E. Gadioli, P.E. Hodgson, *Pre-Equilibrium Nuclear Reactions* (Clarendon Press, Oxford, 1992).
6. P.G. Young, E.D. Arthur, M.B. Chadwick, LA-12343-MS, Los Alamos National Laboratory (1992).
7. D.P. McNabb, UCRL-ID-139906, Lawrence Livermore National Laboratory (1999).
8. J.F. Briesmeister, LA-7396-M-Rev.2, Los Alamos National Laboratory (1986).
9. S.A. Wender, S. Balestrini, A. Brown, R.C. Haight, C.M. Laymon, T.M. Lee, P.W. Lisowski, W. McCorkle, R.O. Nelson, W. Parker, N.W. Hill, Nucl. Instrum. Meth. Phys. Res. A **336**, 226 (1993).
10. J.R. Cooper, J.A. Becker, D. Dashdorj, F.S. Dietrich, P.E. Garrett, R. Hoffman, W. Younes, R.O. Nelson, M. Devlin, N. Fotiadis, UCRL-TR-205760, Lawrence Livermore National Laboratory, (2004).
11. R.O. Nelson et al., *Proc. Int. Conf. Nuclear Data for Science and Technology, Santa Fe, USA, 26 Sept.–1 Oct.*, p. 838 (2004).
12. A.J. Koning, M.B. Chadwick, Phys. Rev. C **56**, 970 (1997).
13. T. Kawano, T. Ohsawa, M. Baba, T. Nakagawa, Phys. Rev. C **63**, 034601 (2001).
14. T. Kawano, Phys. Rev. C **59**, 865 (1999).
15. S. Kunieda, S. Chiba, K. Shibata, A. Ichihara, E.Sh. Sukhovitskii, (in press J. Nucl. Sci. & Technol.) (2007).
16. V. Avrigeanu, P. Hodgson, M. Avrigeanu, Phys. Rev. C **49**, 2136 (1994).
17. R.B. Firestone, *Table of Isotopes* (Eighth Edition, John Wiley & Sons, Inc., 1998).
18. Reference Input Parameter Library, IAEA-TECDOC-1034 (1998).