

$^{175}\text{Lu}(n,n'\gamma)$ gamma-ray production cross section measurements

A. Blanc^{1*}, G. Boutoux², A. Ebran¹, B. Jurado², V. Méot¹, O. Roig¹, C. Theroine¹

¹CEA DAM DIF, F-91297 Arpajon, FRANCE

²Centre d'Etudes Nucléaires de Bordeaux Gradignan (CENBG), Institut National de Physique Nucléaire et de Physique des Particules, (IN2P3), BP 120, Le Haut Vigneau, F-33175 Gradignan, FRANCE

Abstract

Neutron inelastic scattering cross section on Lutetium 175 have recently been measured at various energies between 0.4 and 1.7 MeV using the 4 MV accelerator at CEA DAM-DIF. A High Purity Germanium detector has been used in order to measure the gamma-rays following the (n,n') reaction and thus extract the gamma-rays production cross-sections for low-lying levels transitions. These results have been compared to a TALYS calculation and are generally in good agreement. Results and calculations will be fully discussed for one of the gamma-ray transition at 396.3 keV.

1 Introduction

Fundamental nuclear physics and applications in reactor physics and astrophysics require accurate neutron-induced radiative capture cross section data. CEA DAM-DIF has started in 2007 a program aiming at measuring neutron-induced cross-sections for many *Lu* isotopes [1]. Because of target radioactivity, direct measurements for short-lived nuclei, such as the ^{173}Lu nucleus, are very challenging and does not allow to extract cross sections above a few hundred of keV. Therefore, complementary measurements, using indirect way of determining information for cross sections, are needed.

In the 70's, Britt and Cramer proposed the surrogate method [3] for the indirect measurement of actinide's fission cross section. In this method, the cross section is determined using an alternate reaction (or surrogate reaction) which proceed through the same compound nucleus as the direct reaction. In 2010, fission cross sections of $^{242,243}\text{Cm}$ and ^{241}Am have been measured by Kessedjian et al. [4] using the surrogate method. From 1 to 6 MeV, the results are consistent with the neutron induced fission cross section measurements, but these results are still discussed [2].

The validity of the surrogate method when applied to the neutron-induced radiative capture has not been demonstrated yet. In this context, the $^{174}\text{Yb}(^3\text{He}, p)$ reaction has been studied in 2010 at Orsay [5]. This reaction is the surrogate of the well know $^{175}\text{Lu}(n,\gamma)$ reaction. The surrogate data present large discrepancies with respect to the neutron-induced data. Indeed, the average spins obtained for the surrogate reaction is 3 to 4 \hbar higher than in the neutron-induced reaction [5]. In order to confirm these results we performed a new experiment in April 2012 using the K150 cyclotron at Texas A&M University which aims to study the $^{174}\text{Yb}(p, d)$ reaction as a surrogate of the $^{175}\text{Lu}(n,\gamma)$.

The Orsay experiment results have been interpreted using optical model calculations. These calculations have been performed at CEA DAM-DIF using the TALYS code [6]. TALYS is a computer code developed by NRG-Petten and CEA DAM-DIF for analysis and evaluation of nuclear reactions which uses a parameter library including ENSDF data [7]. It is built on various models such as pre-equilibrium, direct, fission models and optical model for which potential for neutrons can be read as external input. Recently, Garret et al. [8] used the $^{175}\text{Lu}(n, n'\gamma)$ reaction in order to accurately measure rotational bands and isomeric states in ^{175}Lu . Below 800 keV, most of ^{175}Lu low-lying levels are very well know. Therefore, for the calculations, all these levels have been included as discrete levels and a continuum was used above 800 keV. The optical model potential used for neutrons was evaluated on $n + ^{181}\text{Ta}$ data [9] and was adjusted to reproduce the ^{175}Lu total cross section [10].

*Corresponding author, currently at Institut Laue-Langevin, email: blanc@ill.fr.

In the present work we measured the angle integrated gamma-rays production cross sections from the $^{175}\text{Lu}(n, n'\gamma)$ using a High Purity Germanium detector (HPGe). Results and TALYS calculation have been compared in the 0.4 to 1.7 MeV incident neutron energy range. The Sec. 2 and 3 respectively detail the experimental setup and the data analysis. Results and discussions are presented in detail in Sec. 4 for the gamma-ray transition at 396.3 keV.

2 Experimental setup

The $^{175}\text{Lu}(n, n'\gamma)$ measurement has been performed at the 4 MV accelerator at CEA DAM-DIF. The 4 MV accelerator produces a pulsed proton beam with a 400 ns period which impinges a titanium-tritium target (945 $\mu\text{g}/\text{m}^2$). The neutrons are produced by the $T(p, n)^3\text{He}$ reaction which emits nearly mono-energetic neutrons for which the energy directly depends on the θ_{neut} angle between the proton beam axis and the direction of the neutron emission [11]. Actually, at a fixed θ_{neut} angle the neutron energy distribution is driven by the protons energy loss inside the titanium-tritium target. This energy distribution can be calculated from the proton energy loss per length unit in Titanium provided by SRIM [12]. For instance, at zero degree, a proton beam of $E_p = 2500$ keV produces neutrons in the 1640 to 1720 keV range with a 1673 keV mean energy. This energy distribution is narrow enough that the neutron are considered nearly mono-energetic.

Since the ^{175}Lu is the most abundant isotope (97.41%), $\sim 30\text{g}$ of a Lutetium oxide powder was compressed inside polyethylene cylinder (3 cm in diameter and length) and used as a ^{175}Lu target. Its mass and volume were accurately measured and it contained 9.663×10^{22} ^{175}Lu nuclei, the uncertainty being negligible. The sample was placed, at zero degree, ~ 10 cm from the neutron production target. The gamma-ray production cross-sections were thus measured at six neutron mean energies: 458, 594, 682, 789, 1155 and 1673 keV.

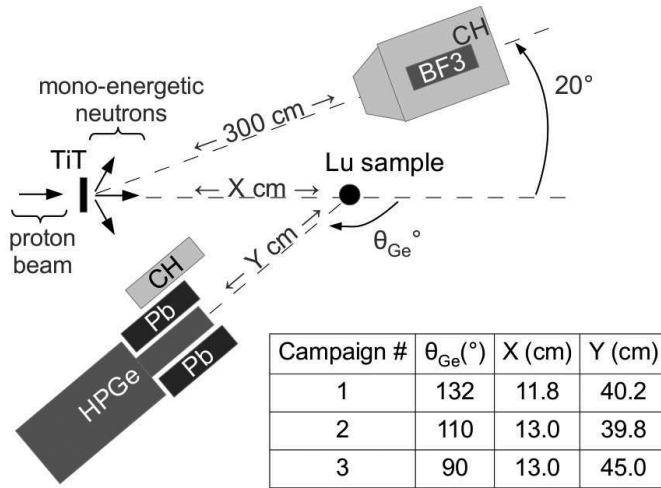


Fig. 1: Schematic view of the experimental setup (not to scale).

The experimental setup consists of a 40% HPGe detector and a BF3 proportional counter for neutron fluence calculation (see Fig. 1). The Germanium detector was placed at a θ_{Ge} angle with respect to the proton beam axis. Three measurement campaigns, at three θ_{Ge} angle (90°, 110° and 132°) were performed in order to extract gamma-rays angular distribution. The θ_{Ge} angle uncertainty was estimated to be $\pm 1.5^{\circ}$. The exact positions of the Lu sample with respect to titanium-tritium target as well as the

position of the HPGe detector with respect to the sample depends on the measurement campaign and are reported Fig. 1. Note that, in order to reduce gamma-ray background, the Germanium detector was shielded with ~ 10 cm of lead. A set of polyethylene was added in order to prevent damage on the Germanium Crystal due to fast neutrons coming from the neutron production target. An accurately calibrated BF3 counter, placed at 3 meters from the neutron production target, at 20 degrees with respect to the proton beam axis allows precise measurement of the neutron fluence in the sample.

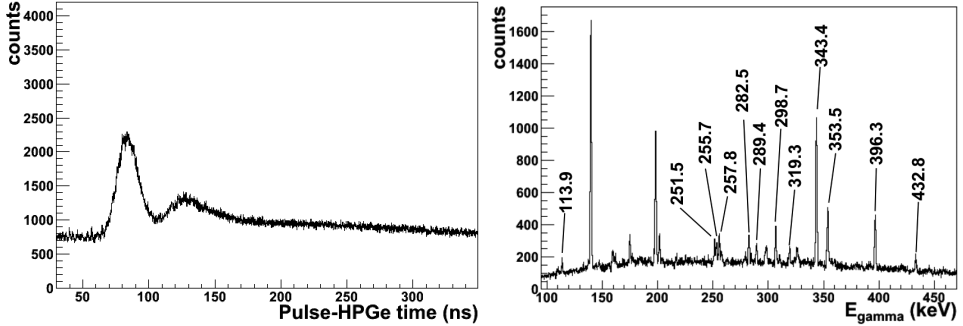


Fig. 2: (left) TAC spectrum for $E_n = 1155$ keV and $\theta = 110^\circ$. (right) Energy spectrum for $E_n = 1155$ keV and $\theta = 110^\circ$ gated on gamma-rays peak (see text for more details). All observed gamma-rays from $^{175}\text{Lu}(n,n'\gamma)$ have been labelled.

Events which are not in time with gamma-rays from the $^{175}\text{Lu}(n,n'\gamma)$ reaction are considered to be uncorrelated background. They mainly come from radioactive elements or thermal neutron capture and their effect can be significantly reduced using the time of flight (TOF) technique. As mentioned above, the proton beam is pulsed and can be used as a START for a time measurement. We thus used a Time Amplitude Converter (TAC) electronic module in order to measure the time between a pulse and a Germanium signal. The Fig. 2 (left) shows a typical TAC spectrum. The first peak at ~ 85 ns is populated by gamma-rays which first hit the Germanium detector. The second peak at ~ 130 ns with a long tail is due to the neutrons. The flat background is due to uncorrelated events. The Germanium detector time resolution was measured and found to be ~ 10 ns. This value is too low to distinguish the prompt gamma-rays (from the neutron production target) from the signal coming from the $^{175}\text{Lu}(n,n'\gamma)$ reaction. The width of the gate used to select the signal has been determined using the delayed ^{175}Lu transition at 396.3 keV and was estimated to be 10 half time, i.e. 32.8 ns. Fig. 2 (right) shows a typical energy spectrum gated on signal. 12 gamma-rays from neutron inelastic scattering on ^{175}Lu have been identified: 113.9, 251.5, 255.7, 257.8, 282.5, 289.4, 298.7, 319.3, 343.4, 353.5, 396.3 and 432.8 keV. The other gamma-rays observed on Fig. 2 (right) mostly come from neutron capture on the Germanium detector.

Experimental setup performances have been measured using calibrated ^{241}Am , ^{133}Ba and ^{152}Eu gamma-ray sources. The energy resolution of the Germanium detector was found close to 2 keV at 1.4 MeV. The setup efficiency at the full energy peak was measured at various gamma-ray energies. Measurements have been compared with a simulation performed using the GEANT4 software [13]. They are in very good agreement above 250 keV [14].

3 Data analysis

TALYS provides angle integrated gamma-rays production cross section for all gamma-rays transitions from discrete levels. In order to compare calculations with measurements, we need to calculate the angle

integrated cross section from the measured gamma-ray yields. To do so we first need to extract from the data the differential production cross section at a θ_{Ge} angle. It was calculated from the following expression:

$$\frac{d\sigma}{d\Omega}(\theta_{Ge}) = \frac{N_{\gamma}(\theta_{Ge})}{\epsilon \cdot N_{nucl} \cdot \Phi_{neut}} \quad (1)$$

where

- ϵ = correction factor including solid angle subtended by the detector (in *sr* unit),
- $N_{\gamma}(\theta_{Ge})$ = gamma-ray yield corrected for the dead time of the acquisition. The dead time correction depends on the trigger rate and range from 0.78 to 0.97,
- Φ_{neut} = neutron fluence in the Lu sample,
- N_{nucl} = number of ^{175}Lu nuclei in the Lu sample,

The correction factor ϵ was determined using GEANT4 simulations which include the setup efficiency (see Sec. 2) and the gamma-ray attenuation in the Lu sample. It ranges from 3.23×10^{-4} to 4.92×10^{-4} *sr* according to the setup and the gamma-ray energy. Its relative uncertainty has been estimated to 5%. As described in Sec. 2, the neutron fluence in the sample Φ_{neut} was calculated from the measurement of the BF3 detector with an uncertainty better than 5%. Note that the energy and fluence variations of the incident neutrons over the sample due to the $T(p, n)^3He$ reaction have been neglected [11]. The neutron attenuation in the sample has also been neglected because of the very low density of the Lu sample (below $1.9 \text{ g} \cdot \text{cm}^{-3}$).

The angle-integrated gamma-ray production cross section as well as the angular distribution were then obtained for each transition. The gamma-ray angular distribution is symmetric about 90° and can be expressed as a series of even order Legendre polynomials [15,16]:

$$\frac{d\sigma}{d\Omega}(\theta_{Ge}) = \frac{\sigma}{4\pi} \sum_{\nu} c_{\nu} P_{\nu} \cos(\theta_{Ge}) \quad (2)$$

where

- $\nu = 0, 2, 4, \dots$,
- σ = angle integrated cross section,
- c_{ν} = coefficient of the angular distribution
with $c_0 = 1$,
- $P_{\nu} = \nu^{\text{th}}$ order Legendre polynomial.

The range of the ν index is determined by vector momentum coupling conditions, in particular, $\nu < 2 \cdot l_n$, where l_n is the angular momentum transferred by the incident neutron [15]. A calculation performed using the TALYS code shows that in the 0.2 to 1.7 *MeV* range, the neutron transmission coefficient for $l_n = 3$ is negligible with respect to $l_n = [0, 1, 2]$ [14]. Therefore, ν vary in the range $[0, 4]$ and the angular distribution can be well described with up to 4th order Legendre polynomial [?, 14]. The differential cross section has been measured at three different θ_{Ge} angle. It results in a system of three equations 2 with three parameters which can be analytically calculated: σ (the angle integrated cross section) and, c_2 and c_4 (the coefficients of the angular distribution).

Note that the effect of the neutron energy distribution in the Lu sample cannot be neglected. It has been included in the analysis by convolving the expected neutron energy distribution in the Lu sample to the TALYS calculation. This energy distribution is mainly due to:

- the proton energy loss in neutron production target (see Sec. 2) which has be calculated using

SRIM [12],

- the multiple scattering in the Lu sample which has been calculated using MCNP [18].

As a result, the calculated cross section decrease from less than 0.01% to 3.56%, depending on the transition and the neutron energy.

4 Results for the gamma-ray at 396.3 keV

The gamma-rays production cross section were obtained for eight transitions: 251.5, 257.9, 282.5, 298.7, 319.3, 343.4, 396.3 and 432.7 keV. They are all above 250 keV where the setup efficiency is well-reproduced by the GEANT4 simulation (see Sec. 2). All of them have been studied in detail in [14]. For the sake of clarity, only one will be discussed in the present proceeding. Since it has been used to determine the width of the gate used to select signal events (see Sec. 2), the delayed transition at 396.3 keV has been chosen.

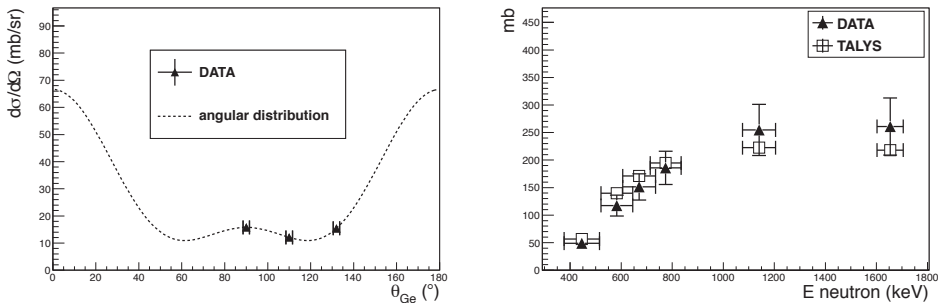


Fig. 3: Results for the gamma-ray at 396.3 keV. (left) Measured differential cross section as a function of θ_{Ge} (full triangles) for $E_n = 1155\text{keV}$. The analytically calculated angular distribution is also plotted (dashed line). (right) Gamma-ray production cross section as a function of the incident neutron energy (full triangles). The TALYS calculation is also plotted (open squares).

The Fig. 3 (left) displays the three measured differential cross sections at three different θ_{Ge} angle for the gamma-ray at 396.3 keV and a 1155 keV mean incident neutron energy. As described in Sec. 3, the angular distribution was analytically calculated from these measurements and the result is also plotted Fig. 3 (left, dashed line). The two "wells" at $\sim 60^\circ$ and $\sim 120^\circ$ illustrates that the effect of the fourth order Legendre polynomial in equation 2 is not negligible. The angle integrated production cross section is then extracted from the angular distribution. The angle integrated cross section as a function of the incident neutron mean energy is plotted Fig. 3 (right, full triangles). As expected the cross section increases with neutron energy increasing and reach a plateau from where the gamma-ray production is mainly driven by the feeding of the corresponding low-lying level. The TALYS calculation convolved with the neutron energy distribution (see Sec. 3) is also plotted Fig. 3 (right, open squares). Results and calculation are in good agreement over the whole neutron energy range studied in the present work.

5 Conclusion

In the present work, gamma-rays production cross section for the $^{175}\text{Lu}(n,n'\gamma)$ reaction have been measured using a 40% HPGe detector. The experiment has been performed at the CEA DAM-DIF 4 MV

accelerator using a nearly mono-energetic incident neutron beam ranging from 0.4 to 1.7 MeV. Differential cross sections have been measured at three HPGe angles with respect to the beam axis. Angular distribution and angle integrated cross sections were then extracted analytically for eight gamma-ray transitions. Optical model calculations has been performed using the TALYS code with a phenomenological potential used during the analysis of the Orsay surrogate experiment [5]. Measurements and calculations are generally in good agreement. These results validate the optical model calculations used for the Orsay experiment. It also validate calculations which, in the same way, will be performed for further experiment aiming at testing the validity of the surrogate method for neutron induced capture cross section measurements in the ^{175}Lu region.

Acknowledgements

The authors thank P. Romain for performing the TALYS calculations and the 4 MV accelerator operations staff for accelerator maintenance and operation.

References

- [1] E. Bauge et al., Eur. Phys. J. A (2012) 48: 113
- [2] P. Romain, B. Morillon, and H. Duarte, Phys. Rev. C 85, 044603 (2012)
- [3] J.D. Cramer & H.C. Britt. Nucl. Sci. Eng. 41, no. 177 (1970)
- [4] G. Kessedjian, B. Jurado, M. Aiche & G. Barreau et al., *Neutron-induced fission cross sections of short-lived actinides with the surrogate reaction method* Physics Letters B, vol. 692, pages 297-301 (2010)
- [5] G. Boutoux et al., *Study of the surrogate-reaction method applied to neutron-induced capture cross sections* Physics Letters B, vol. 712, 319-325 (2012)
- [6] A.J. Koning, S. Hilaire, M.C. Duijvestijn, TALYS: Comprehensive nuclear reaction modeling, in: Proceedings of the International Conference on Nuclear Data for Science and Technology - ND2004, 26 September-1 October 2004, Santa Fe, AIP Conf. Proc. 769 (2005) 1154-1159.
- [7] NNDC On-Line Data Service from the ENSDF database, file revised as of 2011. M. R. Bhat, Evaluated Nuclear Structure Data File (ENSDF), Nuclear Data for Science and Technology, page 817, edited by S. M. Qaim (Springer-Verlag, Berlin, Germany, 1992).
- [8] P.E. Garrett et al., *Rotational bands and isomeric states in ^{175}Lu* , Phys.Rev C 69, 017302 (2004)
- [9] P. Romain and J.P. Delaroche, Proceedings of the Specialist' Meeting on the Nucleon Nucleus Optical Model up to 200 MeV, Bruyères-le-Châtel (1996)
- [10] K. Wisshak, F.Voss, F.Kappeler & L.Kazakov, *Stellar neutron capture cross sections of the Lu isotopes*, Phys.Rev. C 73, 015807 (2006)
- [11] H. Liskien and A. Paulsen, Atomic Data and Nuclear Data Tables 11, 569 (1973).
- [12] <http://www.srim.org/>
- [13] <http://geant4.cern.ch/>
- [14] A. Blanc et al., to be published
- [15] E. Sheldon & D.K. Van Patter, Compound Inelastic Nucleon and Gamma-Ray Angular Distributions for Even- and Odd-Mass Nuclei Review of Modern Physics, vol. 38, no. 1, 143 (1966)
- [16] M.K. Banerjee & C.A. Levinson, Direct interaction theory of inelastic scattering Part I Annals of Physics:2, 471-498 (1957)
- [17] Y.J. Ko et al., Thulium-169 neutrons inelastic scattering cross section measurements via the $^{169}\text{Tm}(n, n'\gamma)$ reaction, Nuclear Physics A, 679, 147-162 (2000)
- [18] <http://mcnpx.lanl.gov/>