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Isomeric ratio measurements for the radiative neutron capture ^{176}Lu(n,γ) at DANCE

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Abstract. The isomeric ratios for the neutron capture reaction ^{176}Lu(n,γ) to the $J^\pi = 5/2^-$, 761.7 keV, $T_{1/2} = 32.8$ ns and the $J^\pi = 15/2^+$, 1356.9 keV, $T_{1/2} = 11.1$ ns levels of ^{177}Lu, have been measured for the first time with the Detector for Advanced Neutron Capture Experiments (DANCE) at the Los Alamos National Laboratory. These measured isomeric ratios are compared with TALYS calculations.

1. Introduction

Neutron capture cross sections are of high interest in nuclear astrophysics to investigate the s-process in which the synthesis of heavy elements is dominated by neutron-induced reactions. In this context, partial cross sections feeding the ground states or isomers are particularly crucial in certain cases of the s-process nucleosynthesis [1–3], or of astrophysical environments such as neutron stars or supernovae where reactions on the isomeric states can occur [4,5].

From the first studies on isomeric states [6,7], the isomeric ratio, defined here as the ratio of isomeric over total (n,γ) cross sections, was always seen as a pertinent parameter to characterize the γ-ray cascade following the decay of the compound nucleus state.

Several methods have been used to measure isomeric ratio in neutron capture reactions: the activation method when the lifetime of the capture reaction product is sufficiently long [8–10], the calorimetry method measuring the total γ energy when excitation energy of isomeric states is sufficiently high compared to the detector energy resolution [11,12] and the method combining time and calorimetry for short lifetimes and high excitation energy of isomeric states [13].

In this paper, we describe a new method using the Detector for Advanced Neutron Capture Experiments (DANCE) array (Los Alamos National Laboratory) driven by a digital data acquisition system. We measured the isomeric ratios for the two short lifetime isomers of ^{177}Lu formed by radiative capture reactions on ^{176}Lu which is one of the two natural isotopes with the highest ground state spin ($J^\pi = 7^-$).

2. Experimental setup

2.1. The DANCE array

The experiment was conducted at the Los Alamos Neutron Science Center (LANSCE) of the Los Alamos National Laboratory. The neutron beam is produced by spallation reactions of the 800-MeV pulsed proton beam impinging on a moderated tungsten target. The DANCE array is located at the end of a 20.25 m long flight path, named FP14. It is composed of 160 barium fluoride (BaF_2) scintillators. This nearly 4π detector is used to measure the total γ-ray energy coming from the neutron capture on the target located at its center. This one is placed in a vacuum beam tube surrounded by a 6 cm thick ^{6}LiH neutron-scattering shield. The pulses from the BaF_2 crystals are digitized with a sampling rate of 500 MHz (2 ns per point) and a resolution of 8 bits [14].

The DANCE array provides the γ-sum energy E_Σ, the multiplicity of crystal hits and the γ-rays energies and times from each crystal for each γ-cascade following the neutron capture. To take into account the Compton effect inside the ball array, a cluster of hits is defined when at least one neighboring crystal is hit. Cluster multiplicity M and cluster γ-rays energies E_γ determined for each γ-cascade are also available. A cluster time is also defined as the mean of all the crystal times which compose this cluster.

2.2. The targets

The ^{176}Lu target is unique with an isotopic enrichment of 99.995%, a mass of 0.542 ± 0.022 mg/cm^2 and a deposit diameter of 7 mm on a 1 μm aluminized Mylar backing. To evaluate the background due to neutron scattering on the target backing, we used a blank target which is composed of the same backing material as the Lu target.

3. Data analysis

The isomeric cross-section ratio R_{iso} of an isomer located at an excitation energy E_{iso} is defined as the ratio of the isomer production cross section σ_{iso} and the total (n,γ) cross section and can be determined from the measurement using:

$$R_{iso} = \frac{\sigma_{iso}}{\sigma_{tot}(n,\gamma)} = \frac{N_{iso}^{exp}\,\epsilon_{casc}}{N_{casc}^{exp}\,\epsilon_{iso}} \qquad (1)$$

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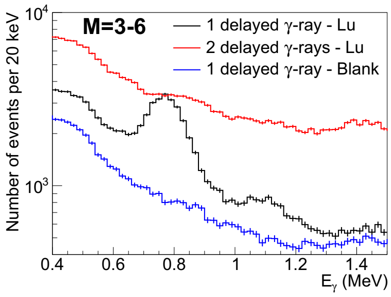


Figure 1. Delayed γ-ray spectra obtained with the DANCE array for $M = 3$ to $M = 6$ cluster multiplicities. Spectra with only one delayed γ-ray for the Lu and the blank targets and two delayed γ-rays for the Lu target are plotted.

where N^{exp}_{iso} is the number of detected isomers, N^{exp}_{casc} is the number of detected cascades, ϵ_{casc} is the cascade detection efficiency and ϵ_{iso} is the isomer detection efficiency. In this section, we will describe how the N^{exp}_{iso} and N^{exp}_{casc} are determined. The simulations used to obtain the ϵ_{casc} and ϵ_{iso} efficiencies are described in Sect. 4.

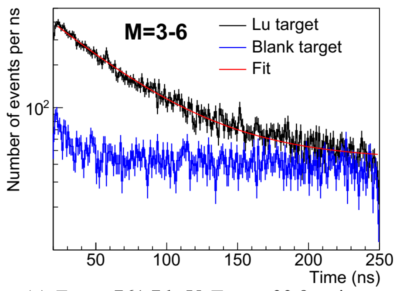
3.1. Selection of the isomers

Our method consists in a careful study of γ-cascades using the time information of each γ-ray of the cascade. In a large coincidence window of 250 ns defined in the off-line analysis, all γ-rays following the neutron capture are recorded and define a capture event. The first γ-rays in this window arriving in a 20 ns width window define the prompt γ-cascade. The other γ-rays are described as delayed γ-rays. The duration of the prompt cascade is defined as the mean time of the individual clusters which compose this cascade. With our method, the decays of the $T_{1/2} = 155\,\mu$s and the $T_{1/2} = 160.4$ d isomers cannot be measured. As their direct feeding are expected to be very low, the influence of these isomers on the total number of γ-cascades can be neglected. The decay of the $T_{1/2} = 130$ ns isomer cannot be also measured as its decay proceeds by the emission of a γ-ray with an energy below the DANCE detection threshold which is 150 keV.

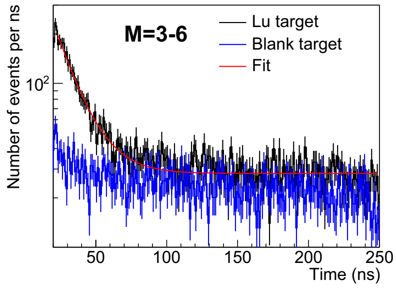
The selection of events with a particular number of delayed γ-rays highlights particular isomer decays. This selection is illustrated in Fig. 1 for cluster multiplicities summed from $M = 3$ to $M = 6$ without any selection in the cascade sum energy. The multiplicity M is the total cluster multiplicity of the cascade which includes the prompt and delayed γ-rays. In Fig. 1, the following γ-ray spectra are presented: the spectrum with only one delayed γ-ray for the Lu and blank targets and the spectrum with two delayed γ-rays for the Lu target.

The spectrum which represents the events with only one delayed γ-ray ($M - 1$ prompt γ-rays) exhibits two peaks located at 762 keV and around 1.1 MeV. These peaks are not present in the case of the blank target which indicates that they are associated with the ^{177}Lu decay. To identify these peaks, energy gates taking into account the BaF_2 energy resolution, are defined around the full energy peaks at 762 keV and 1.1 MeV on the spectrum with one delayed γ-ray.

In Fig. 2, the energy-gated time spectra which represent the difference between the time of the prompt γ-cascade and the delayed γ-ray are plotted for the



(a) E_{iso} = 761.7 keV, $T_{1/2}$ = 32.8 ns isomer



(b) E_{iso} = 1356.9 keV, $T_{1/2}$ = 11.1 ns isomer

Figure 2. Time spectra obtained by applying energy gates on energy spectra with one delayed γ-ray for the Lu and blank targets. The exponential fits used to obtain the isomer population and lifetime are also plotted.

Table 1. Results from the time decay fits performed for the studied isomers.

Isomer	T (ns)	A_0 (counts/ns)	B (counts/ns)
761.7 keV	35.0(9)	522.8(115)	43.1(13)
1356.9 keV	10.8(5)	750.3(622)	28.2(5)

Lu and blank targets for multiplicities summed from $M = 3$ to $M = 6$. As we select capture events with a low number of delayed γ-rays, those where the prompt cascade is not in the first 20 ns of the large coincidence window (250 ns) are rejected. Due to the selection of a high multiplicity prompt cascade, the energy-gated time spectra are not contaminated by the low-multiplicity background events which are removed. The isomer exponential decays are clearly visible for the Lu target whereas the time spectra are flat for the blank target except for the shortest time when the influence of the prompt cascade can be observed. An exponential fit, $f(t) = A_0 \exp(-\frac{\ln 2}{T} t) + B$ is performed to extract the lifetimes and the number of detected isomers for this multiplicity range. The fit parameters are reported in Table 1. According to the fit results, the peaks located at 762 keV and 1.1 MeV are associated with isomers with lifetimes of $T_{1/2} = 35.0 \pm 0.9$ ns and $T_{1/2} = 10.8 \pm 0.5$ ns, respectively. In the nuclear table from [15], these two isomers are identified as the $J^\pi = 5/2^-$, $T_{1/2} = 32.8 \pm 2.4$ ns level located at 761.7 keV and the $J^\pi = 15/2^+$, $T_{1/2} = 11.1 \pm 1.0$ nslevel located at 1356.9 keV respectively. In this database, the $T_{1/2} = 11.1$ ns isomer is fed by another isomer located at 1437.9 keV whose lifetime is not accurately known: $T_{1/2} < 13$ ns. As we have not succeeded in fitting the decay curve of Fig. 2b with two lifetimes, either the lifetime of the 1437.9 keV isomer is about a few ns or it is weakly populated. The number of

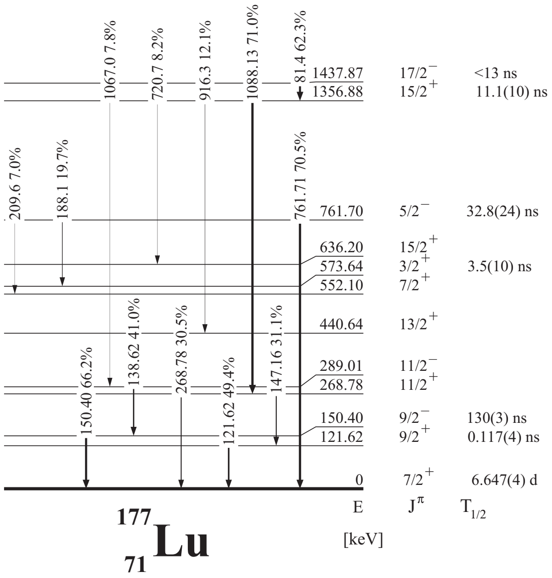


Figure 3. Partial level scheme of the ^{177}Lu [15,16]. The absolute γ intensities are reported.

detected isomers N_{iso}^{exp} is deduced from these fits by:

$$N_{iso}^{exp} = \frac{A_0 T}{\ln 2\, I_{1\gamma}} \quad (2)$$

where $I_{1\gamma}$ is the probability that the isomer decays by only one delayed γ-ray taking into account the DANCE detection threshold and the level scheme which is presented in Fig. 3.

The de-excitation of the $T_{1/2} = 32.8$ ns isomer to the ground state proceeds mainly by the emission of only one γ-ray at 761.7 keV (γ intensity of $I_\gamma(761.7) = 70.5 \pm 2.8\%$). The full energy peak located at 762 keV is thus clearly visible in the spectrum with only one delayed γ-ray. Thus, we have: $I_{1\gamma} = I_\gamma(761.7) = 70.5 \pm 2.8\%$ for the 761.7 keV isomer.

The $T_{1/2} = 11.1$ ns isomer decays mainly by the emission of a γ-ray at 1088.1 keV ($I_\gamma(1088.1) = 71.0 \pm 4.4\%$ if the 1206 keV γ-ray intensity is considered as negligible)[1] toward the $J^\pi = 11/2^+$ level located at 268.8 keV. The de-excitation of the latter proceeds by the emission of a γ-ray of 147.2 keV to the $J^\pi = 9/2^+$ 121.62 keV level or by the emission of a γ-ray of 268.8 keV to the ground state. As the internal conversion coefficients associated with these transitions have not been measured, we have calculated them with the BrIcc code [17]: $\alpha_e(147.2) = 1.13\,(4)$ and $\alpha_e(268.8) = 0.1071\,(15)$. The deduced γ intensities are: $I_\gamma(147.2) = 31.1 \pm 0.7\%$ and $I_\gamma(268.8) = 30.5 \pm 0.9\%$. As the energy threshold applied on the DANCE crystal energy is 150 keV, the 147.2 keV γ-ray cannot be measured. In this case, the majority of the decay of the $T_{1/2} = 11.1$ ns isomer is seen as an unique delayed γ-ray emission. Due to the low energy resolution of the BaF_2 detectors, the peak at 1088.1 keV is blended with the peak at 1067.0 keV also associated with the decay of the $T_{1/2} = 11.1$ ns. As shown in the Fig. 3, the emission of the 1067.0 keV γ-ray is followed by the emission of two γ-rays which are below the DANCE detection threshold (γ-ray at 121.6 and 150.4 keV). By taking into account the DANCE detection threshold, the decay of the $T_{1/2} = 11.1$ ns isomer is mainly detected with only one delayed γ-ray. Thus we have: $I_{1\gamma} = I_\gamma(1088.1)(1 - I_\gamma(268.8)) + I_\gamma(1067.0) = 56.8 \pm 4.2\%$ for this isomer.

[1] Its intensity is not accurately known and its placement in the level scheme is uncertain [15].

3.2. Number of detected cascades

The number of detected cascades N_{casc}^{exp} is summed for cluster multiplicities ranging from $M = 3$ to $M = 6$ because the background is too high for multiplicities less than 3. It is determined, for each multiplicity, by integrating the total energy E_Σ spectrum on a wide energy range around the neutron separation energy $S_n = 7.073$ MeV: $E_\Sigma \in [3.5; 7.5]$ MeV. The case where there is only one cluster hit in the first 20 ns of the large coincidence window of 250 ns (total cluster multiplicity M) corresponds to a background event and is corrected. In this case, the sum energy is calculated by summing the residual γ-ray energies of the events and is reported in the sum energy spectrum corresponding to the $M - 1$ multiplicity. The background spectrum is subtracted before the integration. This one is obtained in the same way for each multiplicity with the blank target.

4. Detection efficiencies

The cascade and isomer detection efficiencies are calculated with GEANT4 [18] simulations of the DANCE detector [19] and with input from theoretical γ-cascades calculated with our Monte-Carlo code EVITA, which is based on the Hauser-Feshbach formalism. This code is able to reproduce a γ-cascade event using all available information for each nucleus involved in the capture reaction. The level scheme and neutron transmission coefficients used as inputs in EVITA are calculated by the TALYS code [20] (version 1.4) and optimized on the specific available information on the desired isotope and reaction. To reproduce the experimental γ-ray spectra, the addition of a resonance with a Lorentzian shape in the Photon Strength Function (PSF) is required as previously suggested by Bečvář et al. [21]. The parameters of this resonance are: $E = 4.25$ MeV, $\Gamma = 2.0$ MeV and $\sigma = 3.75$ mb. The electromagnetic nature of this resonance cannot be determined with our DANCE data.

The cascade detection efficiency ϵ_{casc} is defined as the ratio of the number of detected cascades for $M \in [\![3, 6]\!]$ in the simulated DANCE array, which is calculated by integrating the total energy spectrum in the energy range [3.5–7.5] MeV, and the total number of initial cascades calculated with EVITA. Our cascade simulations are performed with a $M1$, $E1$ or $E2$ resonance added in the corresponding PSF. As the electromagnetic nature of the resonance is not determined, ϵ_{casc} is calculated as a mean and standard deviation over the results obtained with the different resonances. We obtain: $\epsilon_{casc} = 54.9 \pm 1.1\%$.

The isomer detection efficiency ϵ_{iso} is also calculated with GEANT4 simulations using EVITA. It is defined as the ratio of the full energy peak integral of the γ-rays used to evaluate N_{iso}^{exp} summed over the

Table 2. Isomeric ratios calculated with TALYS compared to the experimental ones.

Isomer	Added resonance	Calc. R_{iso} (%)	Exp. R_{iso} (%)
$E_{iso} = 761.7$ keV, $T_{1/2} = 32.8$ ns	Ø	4.1	10.5 ± 0.6
	$M1$	5.7	
	$E1$	5.0	
	$E2$	6.3	
$E_{iso} = 1356.9$ keV, $T_{1/2} = 11.1$ ns	Ø	2.6	4.8 ± 0.6
	$M1$	2.0	
	$E1$	2.0	
	$E2$	1.6	

multiplicities $M \in [\![3, 6]\!]$ and the total number of the same γ-rays in the initial cascades calculated with EVITA. In the same way as for ϵ_{casc}, ϵ_{iso} is an average over the results of the simulations obtained with the different types of added resonances. We obtain: $31.7 \pm 0.5\%$ and $38.0 \pm 0.6\%$ for the 761.7 keV and 1356.9 keV isomers respectively.

5. Results and discussions

The obtained experimental isomeric ratios are summarized in Table 2. They are compared with TALYS calculations for the different resonances added in the PSF. Large discrepancies between experimental and evaluated isomeric ratios can still be observed regardless of the addition and the nature of the added resonance. The agreement between experimental and calculated isomeric ratios is improved by adding an $E2$ resonance for the 761.7 keV isomer. On the other hand, it is better without any resonance in the case of the 1356.9 keV isomer.

6. Conclusion and perspectives

For the first time, isomeric ratios for two isomers of the ^{177}Lu compound nucleus, at 761.7 keV ($J^{\pi} = 5/2^{-}$, $T_{1/2} = 32.8$ ns) and 1356.9 keV ($J^{\pi} = 15/2^{+}$, $T_{1/2} = 11.1$ ns) have been obtained from a TOF experiment at DANCE integrated over the neutron energy range: 8 eV–100 keV.

The experimental isomeric ratios were compared with TALYS calculations. Large discrepancies were observed between the data and the calculations. Different parameters and models were unsuccessfully tested to reproduce the experimental results. The level scheme, which is a key ingredient in this kind of calculations needs to be improved in order to better reproduce the data.

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