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Direct measurement of the inelastic neutron acceleration by ^{177m}Lu

V.Méot[1,a], O. Roig[1], B. Rossé[1], P. Morel[1], J-M. Daugas[1], D.Doré[2], A. Letourneau[2], A. Menelle[3]

[1] CEA DAM DIF, F-91297 Arpajon, FRANCE
[2] CEA DSM IRFU, F-91191 Gif-sur-Yvette, France,
[3] CEA/CNRS LLB, F-91191 Gif-sur-Yvette, France

Abstract. The inelastic neutron acceleration (INNA) cross section on the long-lived isomer state of ^{177m}Lu has been measured from a new isomeric target using a direct method. The detection of high energy neutrons has been performed using a specially designed setup and a cold neutron beam at the ORPHEE reactor facility in Saclay.

1 Introduction

Super-elastic neutron scattering, also called inelastic neutron acceleration (INNA), occurs during the collision of a neutron with an excited nucleus. In this reaction, the nucleus transfers some of its excitation energy to the scattered neutron. Predicted [1] since 1959, this process was observed twenty years later on two isomeric nuclei [2] [3]. The effect of this neutron induced de-excitation is known in nuclear astrophysics to take place in the s-process nucleosynthesis [4] . Due to the usually high cross section of the thermal neutron interaction, such a process could be used to induce a fast de-excitation of an isomer. The 160-day $23/2^-$ isomer in ^{177m}Lu (Ex = 970 keV) is a good candidate to observe the inelastic neutron acceleration reaction. The production [5] of an easy to handle target has been undertaken several times at the Laüe Langevin Institute (ILL). The thermal INNA reaction cross section for ^{177m}Lu has been deduced indirectly from burn-up [6] and capture cross sections [7] to be a high value, (258±58 b), enabling direct detection of the high energy neutrons. In this paper, we report on experimental results on the direct measurement of high energy neutrons in the reaction between the long-lived isomer of ^{177m}Lu and cold neutrons at the ORPHEE reactor facility. From the previously published value of the INNA cross section and from these measurements, we discuss the relevance of these latter results.

2 Experimental method

2.1 The ^{177m}Lu Target

10.75 mg natural Lutetium foil (25μm thickness) was irradiated at the High Flux Reactor (HFR) at the Laüe Langevin Institute (ILL) in Grenoble (France). The V4 port was used to take advantage of the maximum neutron flux, 1.5 10^{15} neutrons.cm^{-2}.s^{-1}. The ^{natural}Lu foil was put inside an aluminium capsule with a beam monitor sample of AlCo and was irradiated during 6 days. The beam monitor was used to determine the neutron flux at the V4 port during the irradiation time. After a 150 days cooling time the irradiated foil was put inside a thin pure aluminum envelope (20μm thickness). The number of the (1.03±0.04) 10^{14} isomers was accurately measured by performing a γ-spectroscopy at the time of the experiment. A similar non-irradiated natural Lutetium foil from the same sample used for the isomer target was used as blank target.

[a] e-mail: vincent.meot@cea.fr

2.2 The ORPHEE facility

The 14 MW reactor ORPHEE is made up of a very small size highly enriched in 235 Uranium core, surrounded by heavy water reflector tank to get a good thermal flux, 3 10^{14} n.cm^{-2}.s^{-1}. Several cold beams are extracted along "neutron guides" emerging from the reactor building. For this experiment, we used the G3-2 point located about 20 m from the reactor core. The neutron guides are composed of multi-layers of nickel-titanium mirrors and are slightly curved in order to suppress the fast neutrons and gamma-ray background from the core. The internal section of the G3-2 neutron guide is 25×50 mm^2 giving a neutron flux of 1.5×10^9 n.cm^{-2}.s^{-1} for a mean energy of 4.5 meV and with an angular spreading of 0.4°. In order to decrease the background and to increase the detection efficiency, the neutron beam was collimated to a diameter of 2 cm. The collimator was made up of a 1 cm thickness boron carbide neutron absorber, followed by a 1 cm thickness CH_2 layer and a 2 cm thick lead gamma attenuator. It is placed at 1.8m upstream of the target. A second collimator, made of a 1 cm B_4C and a 5 cm lead layers, is placed as close as possible to the target in order to eliminate neutrons scattered on the first collimator.

2.3 Neutron detector

The main difficulty in this experiment is to overcome the low signal to background ratio. Two background sources have to be considered. The first one is due to scattered cold neutrons. To limit the count rate a dedicated chamber has been built to work under vacuum. The second one is due to photons from neutron captures and the gamma activity of the target itself. Because of its low sensitivity to gamma-rays, the choice of the 10 bar pure ^{3}He proportional counter was done. Its outer diameter is 1 inch and its active length is 25 cm. Despite the high photon flux, the neutron detection is unaffected because the deposited energy is small and associated with a very low efficiency. Moreover the neutron detection is obtained via the exothermic reaction: n + ^{3}He $\leftrightarrow$ ^{3}He + p, whose Q value is 764 keV. Tests with a very intense^{60}Co source showed that the thermal neutron peak located at 764 keV was not affected for photon fluxes up to 10^7 ph/s on the detector. To detect high energy neutrons, a slowing down neutron counter was used. The neutron array consists of a cylindrical polyethylene moderator combined with twelve proportional ^{3}He counters placed inside the moderator. Fast neutrons produced in the target scatter many times in the moderator and quickly slow down by elastic scattering to the thermal neutron energy. Then, the ^{3}He detectors have a large efficiency to detect these thermal neutrons. The inner part of the vacuum chamber is shielded by a 2 mm cadmium layer to suppress the neutrons scattered by the target. An outside 1mm layer of cadmium covers the whole cylindrical polyethylene moderator in order to stop all thermal neutrons coming from the background or cold neutrons scattered by the collimator. The cadmium layer lets come high energy neutrons into the polyethylene cylinder where they loss their energy. The neutron array was simulated using MCNP code to get the thermal neutron total efficiency. For neutron energies between 100 keV and 500 keV, which are the expected values for the emitted neutrons, the efficiency is flat and is worth 0.198. A very similar response was obtained using GEANT4. However, our simulation does not take into account the wall effects (only a part of the energy is deposited in the ^{3}He). The correction factor was obtained by measuring and simulating the neutron activity of a known ^{252}Cf source positioned in the target holder. The right efficiency to be used is then 0.18

2.4 Neutron flux determination

In order to obtain the exact neutron flux on the target, the activation of the ^{176m}Lu isomer produced in the radio-active target by ^{175}Lu(n,γ) reaction, was measured just after the irradiation. A LEPS germanium detector was used to measure the 88.361 keV γ-ray from the β-decay of the ^{176m}Lu. The amount of ^{175}Lu present in the isomeric target could be determined with accuracy knowing the initial ^{175}Lu quantity, the neutron flux in V4 at ILL and the combustion cross section. To extract the flux we need to know the value of the ^{175}Lu(n,γ) cross section in the cold G3-2 flux. The reaction rate can be written as:

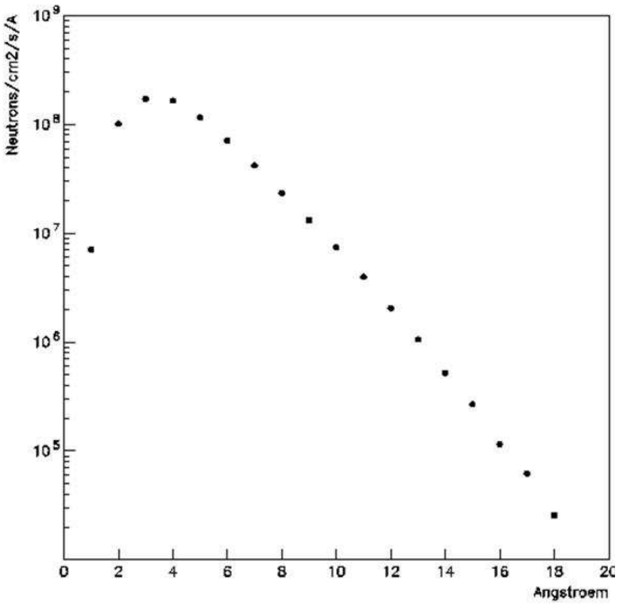

Fig. 1. Neutron flux at the G 3-2 point

$$R = \phi \int_0^\infty S(\lambda) \times \sigma(\lambda) d\lambda \quad (1)$$

where is the neutron flux, previously determined [8] S(λ) the G3-2 neutron spectrum (see figure 1), λ the neutron wavelength and σ the neutron capture cross section. If the capture cross section in the neutron flux range obeys to the 1/v law, the integration is straight forward :

$$\phi \int_0^\infty S(\lambda) \times \sigma(\lambda) d\lambda = \frac{\sigma_0}{\lambda_0} \int_0^\infty S(\lambda) \times \lambda d\lambda \quad (2)$$

where σ_0, 16.2 barns, is the ^{175}Lu(n,γ)^{176m}Lu cross section at thermal energy (λ_0=1.805 A and E_0=0.025 eV). In the cold G3-2 flux, the ^{175}Lu(n,γ)^{176m}Lu cross section is then 40.29 barns. Finally, using this value and the ^{176m}Lu activation measurement, we obtain a neutron flux of 1.88±0.20 10^8 n/cm^2/s on the target.

Using calibrated gold sample activation, the neutron flux was measured several times during the experiment to take into account fluctuations of the reactor thermal power. In addition, the reactor thermal power file was analyzed to check discontinuities.

2.5 Measurement analysis

The measurement consists in counting the thermal neutrons for various configurations. The counting rate with the neutron beam impinging the Lu radioactive target is 4.4±0.2 c/s. The background is composed of various components: the high activity of the target, the noise arising from the collimation of the beam, neutron scattering on the

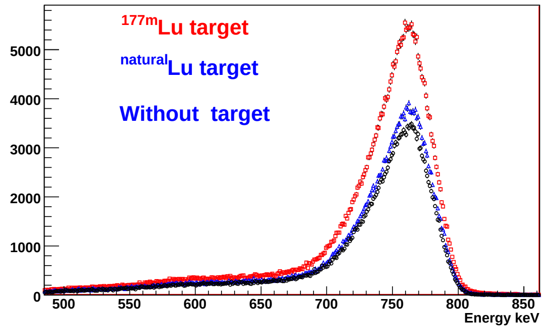

Fig. 2. Response of the Neutron array for various configurations.

target and neutrons from the environment (presence of the reactor, natural radioactivity...). In order to determine the contribution of these backgrounds, we performed different measurements. The true count of thermal neutrons corresponding to the high energy neutrons will be obtained by subtracting all the backgrounds from the full configuration. We present in the following the analysis of all these configurations. When the isomeric target is put inside the chamber with no beam, the counting rate is 1.25 c/s. In order to check the influence of the target radio-activity, we performed a measurement with and without the target in two configurations. The first one consists in counting the thermal neutrons with no beam inside the chamber, the second one in stopping the neutron beam in a B_4C block in front of the target. In both configurations, the difference with and without the isomeric target is less than 0.2 c/s. We controlled the effect of the beam inside the chamber without any target. The counting is then 2.8±0.2 c/s. The contribution of the neutrons scattered on the target was controlled by means of natural lutetium targets. For 6.8 mg, 12.2 mg and 14 mg, the corresponding counting rates is 3.1±0.2 c/s. The incertitude of the measurement comes from the imprecision in replacing the target inside the chamber and/or is due to the variations of the reactor power between the different runs. From all these measurements, the error on the counting rate could be estimated to be 0.2c/s. The figure 2 shows the response function of the neutron array for these different configurations. The true count of thermal neutrons corresponding to high energy neutrons is obtained by subtracting all the background to the full configuration. The result obtained is 1.3±0.2 neutrons/s. The observed signal could be explained neither by the target radioactivity nor the cold neutron scattered by the target. From the knowledge of the neutron flux, the detection efficiency and the number of isomers, the cross-section of the observed signal may be deduced. The analysis is in progress.

3 Conclusion

The inelastic neutron acceleration cross section on the long-lived isomer state of ^{177}Lu has been measured in a cold neutron beam. This result confirms the existence of the phenomenon. The analysis will allow us a comparison with the previous indirect measurement [6].

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