

Evidence for inelastic neutron acceleration by the ^{177}Lu isomer

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The neutron burnup cross section σ_{burnup}^m on the long-lived metastable state of ^{177}Lu has been measured from a specially designed isomeric target. The Maxwellian averaged cross section obtained for this reaction on $^{177}\text{Lu}^m (J^\pi = 23/2^-)$ is $\sigma_{\text{burnup}}^m = 626 \pm 45$ b at the reactor temperature $T = 323$ K. The difference between the burnup cross section and the previously measured capture cross section $\sigma_{n,\gamma}$ clearly shows a possible existence of $^{177}\text{Lu}^m$ deexcitation via (n, n') inelastic neutron acceleration channels. The results are interpreted in terms of a statistical approach using parameters from a deformed optical potential calculation.

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I. INTRODUCTION

Nuclear isomers are long-lived metastable states studied for their ability to both probe atomic nuclei structure and store energy [1–3]. In order to release the isomer energy, many experiments have been devoted to the depopulation of such states [4–7]. Among the processes able to induce deexcitation of nuclear isomers, the inelastic neutron acceleration reaction (INNA) could be more efficient than an electromagnetic process. This reaction, also called neutron superelastic scattering, occurs during the collision of a neutron with an excited nucleus. Thus, the nucleus can partly transfer its excitation energy to the scattered neutron. The effect of this neutron-induced deexcitation is known in nuclear astrophysics to take part in the s-process nucleosynthesis [8–10]. Predicted by Petrov [11] in 1959, the first measurement attempt was initiated in 1969 by Miyano and Morinaga [12]. They determined an upper limit for an exothermic nuclear (n, n') process at thermal neutron energy for $^{148}\text{Pm}^m$ of $\sigma \leq 100$ b. Later, Hamermesh [13] searched for an inelastic scattering of thermal neutrons on $^{123}\text{Te}^m$ and obtained an upper limit of $\sigma = 20$ mb. At the same time, theoretical calculations were made by the group of Petrov and Shlyakhter [14,15]. Up to now, this phenomenon has solely been observed on $^{152}\text{Eu}^m$ and $^{180}\text{Hf}^m$ [16,17] by Kondurov *et al.* In these works, the measured cross sections were 0.28 ± 0.06 and 52 ± 13 b, respectively. Due to the low cross section value and high activity of the $^{152}\text{Eu}^m$ and $^{180}\text{Hf}^m$ targets, energies of outgoing neutrons could not be measured, leaving the underlying mechanisms still unknown.

The 160-day $23/2^-$ isomer in ^{177}Lu ($E_x = 970$ keV) [18–20] is a candidate for observing the inelastic neutron acceleration reaction. Its long half-life allows the production of an easy-to-handle target [21,22]. In Ref. [23] we present a method to measure the INNA reaction cross section using an isomeric target. It involves two types of measurements in an equivalent thermal neutron flux. The first measurement gives

the isomer radiative capture cross section, while the second provides the isomer burnup cross section.

In this work, we report on experimental results on the burnup cross section for the long-lived isomer of ^{177}Lu . From these measurements and from the published value of the radiative capture cross section [24], the INNA reaction cross section for $^{177}\text{Lu}^m$ has been deduced. Finally, we discuss the pertinence of this result.

II. EXPERIMENTAL DETAILS

A. Isomeric target

The $^{177}\text{Lu}^m$ was produced by thermal neutron irradiation of a ^{176}Lu sample. This sample was highly enriched (99.993%) using the mass separator PARSIFAL [21] in Bruy res-le-Ch tel. A 1.25 mg sample was irradiated in the high flux reactor (HFR) at the La ie Langevin Institute (ILL) in Grenoble (France) for 49 days in the V4 irradiation port. The total neutron flux was about $1.5 \times 10^{15} n \text{ cm}^{-2} \text{ s}^{-1}$ with 15% of epithermal neutron. After the irradiation period, the sample was cooled down to remove the ^{177}Lu ground state, which is short lived (6.647 ± 0.004 days) compared to the isomeric state (160.44 ± 0.06 days). $^{177}\text{Lu}^m$ targets were produced by a direct deposit method after a chemical separation to extract lutetium from hafnium. The deposit was performed on $25 \mu\text{m}$ thick titanium backings. Table I summarizes the isomeric target characteristics.

B. Radiative capture cross sections

The capture cross section measurements [24] at thermal neutron energy were performed at two different research nuclear reactors, ORPHEE in Saclay using the p1 port and HFR at ILL using the H9 port. In Table II, the values of the Maxwellian averaged capture cross sections $\sigma_{n,\gamma}(T)$ are presented for both reactors at a reactor temperature $T = 323$ K. These values were obtained from the effective cross section values [24] at $T = 323$ K under the assumption that ORPHEE

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TABLE I. Number of atoms in the $^{177}\text{Lu}^m$ targets used at the date of irradiation period.

Target	\emptyset of Ti backing (mm)	$^{177}\text{Lu}^{\text{gs}}$	$^{177}\text{Lu}^m$	^{176}Lu	^{181}Ta
1	5	5.25×10^8	$(5.83 \pm 0.08) \times 10^{10}$	$\approx \times 10^8$	$\approx 10^{11}$
2	19	2.07×10^9	$(2.30 \pm 0.02) \times 10^{11}$	$\approx \times 10^8$	$\approx 1.3 \times 10^{12}$

and HFR/H9 [25] neutron fluxes are fully thermal. The f factor in Table II is the ratio between the thermal and the epithermal components of the neutron flux for each experiment.

C. Experimental setup for the burnup cross section measurements

According to the Westcott convention [26], a reactor flux can be described by two parameters: temperature and f factor. For two different neutron fluxes with identical parameters, the measured cross sections are equivalent. In order to apply the method described in [23], the measurement of the burnup cross sections must be performed in a neutron flux similar to the one used for radiative capture. Two irradiation facilities are adequate for these measurements in the HFR: the HFR/T4 port where the neutron flux is equivalent to the one of ORPHEE/p1 port, and the HFR/H9 port where the radiative capture cross section has already been measured. The burnup cross section measurements consisted in performing a very precise γ -ray spectroscopy analysis of the $^{177}\text{Lu}^m$ sample decays to determine its activity both before and after an irradiation period with the same high-purity-Ge detector setup. The γ -detection efficiencies were made using a calibrated standard source of ^{152}Eu and ^{60}Co placed at the counting position. Statistical errors on the number of γ -ray events as well as solid angle uncertainties were controlled to be lower than 0.5% to minimize the cross section errors. IRMM (Institute for Reference Materials and Measurements) [27] aluminum monitors loaded at 0.1% with ^{59}Co were irradiated with each sample in order to measure the total neutron fluence.

For each port, two specific setups for the irradiation and γ -ray spectroscopy measurements were used. Inside the HFR/T4 port, target 1 [Table I] was confined in a quartz flask lying inside an aluminum container. Irradiation time was 92 days; cooling time was quite long, 110 days. For γ -counting, the position of the target was located at 600 ± 1 mm from the HP-Ge detector in order to minimize the influence of position uncertainty. A ^{207}Bi source permanently located close to the HP-Ge detector was used to determine the uncertainty on dead time and systematic errors. Inside the HFR/H9 port, the nonconfined target 2 [Table I] was deposited on a titanium backing mounted on a titanium holder. A mechanical carriage

TABLE II. Effective $\hat{\sigma}_{n,\gamma}$ and Maxwellian averaged $\sigma_{n,\gamma}$ radiative capture cross sections at 323 K.

Reactor /port	T (K)	f	$\hat{\sigma}_{n,\gamma}^a$	$\sigma_{n,\gamma} = \hat{\sigma}_{n,\gamma} \sqrt{\pi/2}$
ORPHEE/p1	323	0.0005	418 ± 28 b	370 ± 25 b
HFR/H9	323	0.015	413 ± 60 b	366 ± 53 b

^aReference [24].

transported this target holder into the irradiation position and then into the mini-INCA (INCineration of Actinides), chamber [28] used for the counting. Irradiation time was 3 days, and cooling time was 60 days. Inside the mini-INCA chamber, the distance between the target position and the HP-Ge detector during the γ -counting was 415 mm, and the mechanical carriage guaranteed the repositioning.

III. RESULTS

A. Measurements at HFR/T4 and HFR/H9 ports

For experiments performed at HFR/T4 and HFR/H9 ports, the data analysis was restricted to the seven most intense $^{177}\text{Lu}^m$ γ -rays, $E_\gamma = 112.9, 153.3, 208.4, 228.5, 327.5, 378.5,$ and 418.5 keV. Among these, only nonpolluted γ lines were used. The selection was achieved by comparing the measured $^{177}\text{Lu}^m$ γ -ray intensities per decay and the intensities of literature. Polluting elements were determined in the $^{177}\text{Lu}^m$ γ -line spectrum: ^{228}Ac from the background environment, ^{182}Ta activated from the ^{181}Ta in the target [21], and fission products from a known actinides contamination inside the HFR/H9 port.

In Table III, the γ rays used for the measurements are shown. For the HFR/T4 port, due to the low activity of the $^{177}\text{Lu}^m$ target and the setup environment, 153.3, 208.4, and 327.7 keV γ rays were polluted by the background γ rays of ^{228}Ac . The ^{182}Ta γ rays were not detectable in the γ -ray spectrum after an irradiation period at the HFR/T4 port. For the HFR/H9 port, the mini-INCA chamber was protected with a lead shield against the background environment. Due to an important fluence and a shorter cooling time, the activity of ^{182}Ta , produced from ^{181}Ta contained in the target, caused that the 153.3 and 228.5 keV γ were polluted. Figure 1 shows a sample of the γ -ray spectrum of the $^{177}\text{Lu}^m$ following HFR/T4 and HFR/H9 irradiations.

Figure 2 shows the logarithm of the 378.5 and 228.5 keV activities before and after irradiation periods at HFR/T4 and HFR/H9 ports. The differences observed between the

TABLE III. Energy of the seven most intense γ rays of $^{177}\text{Lu}^m$ ordered by decreasing intensity. Polluted γ rays are indicated with the element. Used γ -rays for the HFR/T4 and HFR/H9 measurements are noted.

Energy (keV):	208.4	228.5	378.5	112.9	418.5	327.7	153.3
HFR/T4	^{228}Ac Used	Used	Used	Used	Used	^{228}Ac	^{228}Ac
HFR/H9	Used	^{182}Ta	Used	Used	Used	Fission products	^{182}Ta

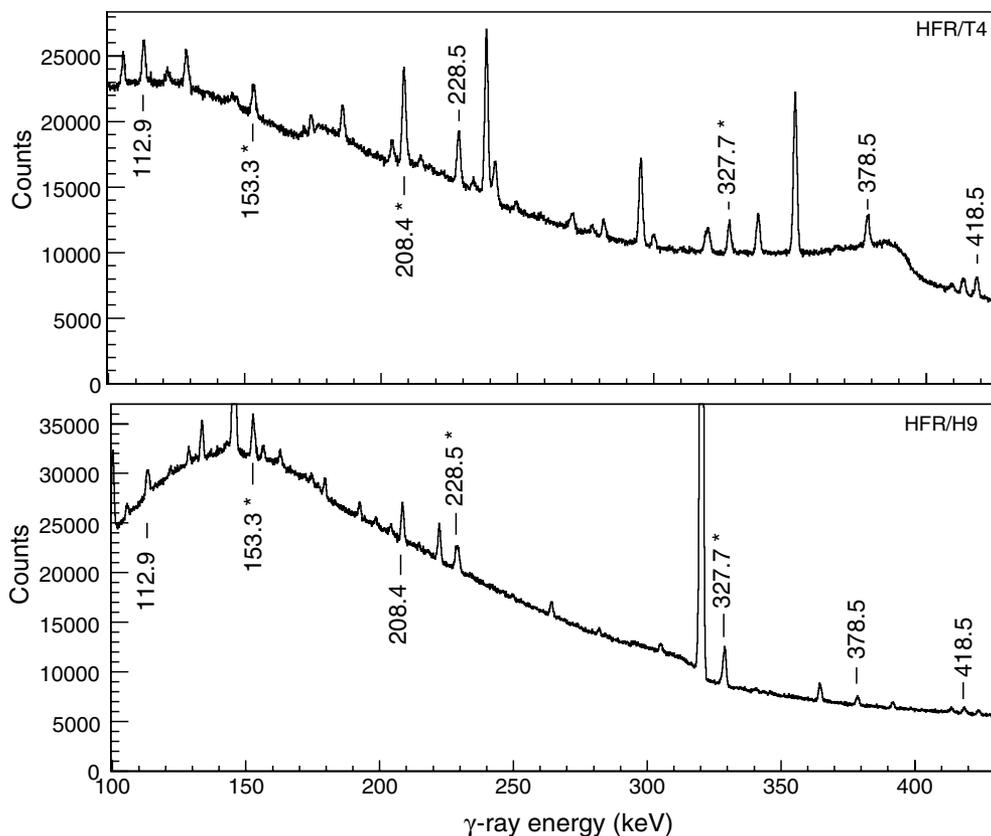


FIG. 1. Sample of the γ -ray spectrum of $^{177}\text{Lu}^m$ after HFR/T4 and HFR/H9 irradiation periods. Among the seven most intense $^{177}\text{Lu}^m$ γ lines, the polluted ones (from ^{228}Ac , ^{182}Ta , and fission product contaminations) are labeled with a star.

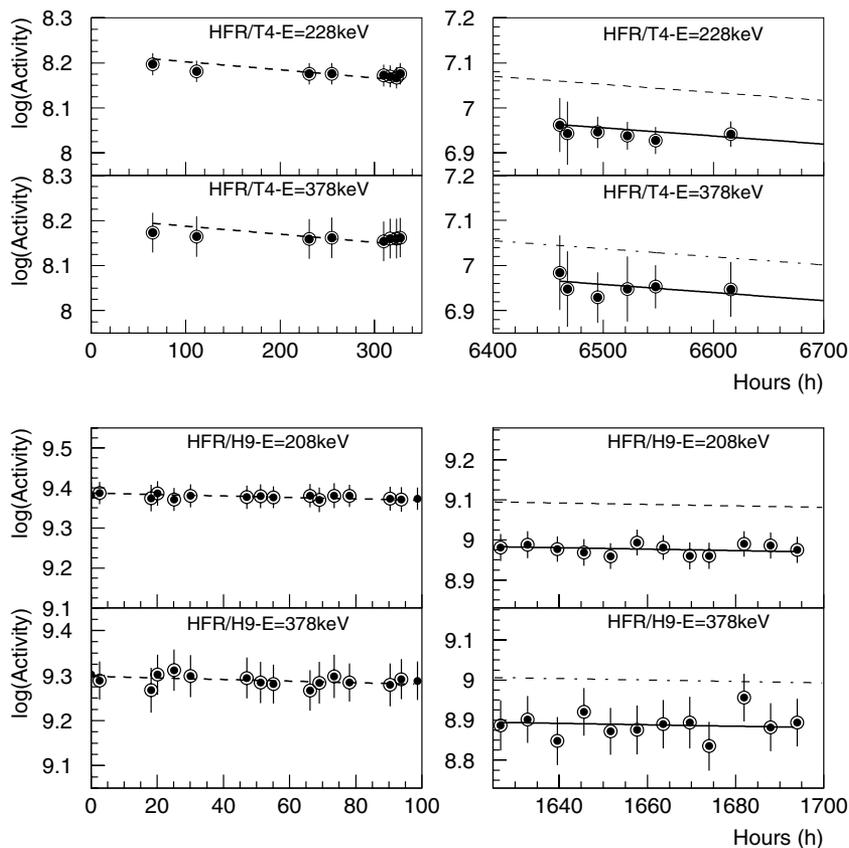


FIG. 2. Logarithms of 208.4, 228.5, and 378.5 keV $^{177}\text{Lu}^m$ γ -ray activities deduced from γ spectroscopy before and after irradiation as a function of time. Error bars include statistical errors of γ counting, time errors for counting, irradiation and cooling periods, and half-life uncertainty (standard deviations). Dotted lines are the logarithms of activities expected without burnup (only $^{177}\text{Lu}^m$ decay).

measured and expected activities after both irradiation periods clearly indicate the $^{177}\text{Lu}^m$ burnup.

The intensities of the nonpolluted adopted $^{177}\text{Lu}^m$ γ -lines were measured, and the burnup cross section value was directly deduced from the weighed average, R , of the ratio R_γ of individual corrected counting rate before and after an irradiation period, defined as

$$R = \Sigma_\gamma \frac{R_\gamma}{(\Delta R_\gamma)^2} \bigg/ \Sigma_\gamma \frac{1}{(\Delta R_\gamma)^2}, \quad \text{where}$$

$$R_\gamma = \frac{N_\gamma^{ic}}{N_\gamma^{fc}} \frac{1 - e^{-\lambda_{177m} t_{\text{counting}}^f}}{1 - e^{-\lambda_{177m} t_{\text{counting}}^i}}$$

where ΔR_γ , t_{counting} , λ_{177m} , N_γ^i , N_γ^f are the error of individual counting rate ratio, the counting time, the $^{177}\text{Lu}^m$ period, and the total count in one γ line before and after an irradiation period, respectively. By using R_γ , the detection efficiency cancels out, as the setup was the same before and after the irradiation period. Then, R does not depend on the efficiency values, and thus systematic errors are reduced. The burnup rate is defined as

$$R_{\text{burnup}}^\gamma = 1 - R_\gamma e^{-\lambda_{177m} t_{\text{decay}}},$$

where t_{decay} is the decay time between the beginning of the counting before and after an irradiation period. Figure 3 shows the burnup rate for each $^{177}\text{Lu}^m$ γ -ray lines and its weighed average for HFR/T4 and HFR/H9 experiments. The dashed line represents the burnup rate under the assumption that the only contributing process in the burnup is the radiative capture.

The burnup cross section was obtained using the formula:

$$\sigma_{\text{burnup}}^m = \frac{-1}{t_{\text{irradiation}} \Phi} \log(R e^{\lambda_{177m} t_{\text{decay}}}),$$

where Φ and $t_{\text{irradiation}}$ are the averaged neutron flux and irradiation time, respectively. The total neutron fluence was measured using an IRMM aluminum monitor loaded at 0.1% with ^{59}Co . The ^{60}Co activity was measured using the detection efficiency obtained from calibrated Co sources and using the $^{59}\text{Co}(n, \gamma)^{60}\text{Co}$ averaged cross section estimated in HFR/T4 or HFR/H9 ports [25].

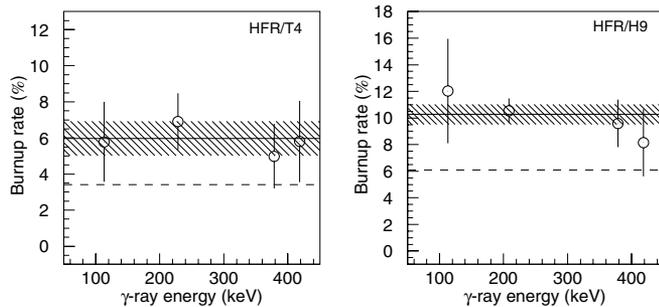


FIG. 3. Averaged burnup rate for four $^{177}\text{Lu}^m$ γ rays (solid line and dashed standard error area), in percentage, compared with individual γ -ray burnup rates for HFR/T4 and HFR/H9 irradiations. Dashed line represents the expected value of the $^{177}\text{Lu}^m$ burnup rate with radiative capture.

TABLE IV. Maxwellian averaged burnup cross sections for $^{177}\text{Lu}^m$ measured at the temperature of 323 K.

Reactor/port	Flux ($n \text{ cm}^{-2} \text{ s}^{-1}$)	f	σ_{burnup}^m (b)
HFR/T4	$(1.24 \pm 0.05) \times 10^{13}$	0.0005	624 ± 93
HFR/H9	$(6.67 \pm 0.32) \times 10^{14}$	0.015	627 ± 51

The values obtained for the averaged burnup cross sections were 624 ± 93 and 627 ± 51 b at the HFR/T4 and HFR/H9 ports, respectively (see Table IV). These quantities correspond to a burnup value of $5.97 \pm 0.95\%$ and $10.19 \pm 0.77\%$, respectively.

The differences between the two neutron energy distributions for the epithermal energy region, inside HFR/T4 and HFR/H9 ports, characterized by the f factor (Table IV), allow us to define the influence of the compound nucleus neutron resonance. Since the measured cross sections are the same, the influence of neutron resonance at epithermal energy is negligible. Thus, the resulting cross section of the burnup reaction on $^{177}\text{Lu}^m$ in a Maxwellian flux is a mean value of these two previous measurements inside the HFR/T4 and HFR/H9 ports:

$$\sigma_{\text{burnup}}^m = 626 \pm 45 \text{ b} \quad \text{at } T = 323 \text{ K.}$$

B. Measurements at HFR/V4 port

A complementary method was also used to measure the burnup cross section for the $^{177}\text{Lu}^m$. It involved in extracting this cross section from a double capture reaction on ^{176}Lu in the highest neutron flux available in the HFR/V4 port. The ^{176}Lu sample was irradiated for 46 days with a neutron flux $\phi(1.53 \pm 0.04) \times 10^{15} n \text{ cm}^{-2} \text{ s}^{-1}$. The neutron fluence seen by the sample was measured using an IRMM AlCo flux monitor. The average neutron flux was obtained using the $^{59}\text{Co}(n, \gamma)^{60}\text{Co}$ average cross section estimated in the HFR/V4 port [25]. Before an irradiation period, the number of atoms N_{176}^0 and the mass of ^{176}Lu , 0.31 ± 0.03 mg, were deduced from the activity, measured by γ -ray spectrometry [29]. The activity was so low that it had to be determined in the Modane underground laboratory (LSM). During irradiation, the production cross sections of $^{177}\text{Lu}^m$ and $^{177}\text{Lu}^{\text{gs}}$, σ_{177m} and $\sigma_{177\text{gs}}$, were estimated at 4.2 and 3153 b, respectively. These values were obtained by averaging the ^{176}Lu experimental cross section [30] with the HFR/V4 neutron energy flux simulated with (MCNP, Monte Carlo N Particles) [25]. The $^{177}\text{Lu}^m$ burnup cross section, σ_{burnup}^m , was determined from the ^{176}Lu mass, the radiative production cross sections of $^{177}\text{Lu}^m, \text{gs}$, and the measured activity, by solving numerically the differential equation

$$\frac{dN_{177m}}{dt}(t) = N_{176}^0 e^{-(\sigma_{177m} + \sigma_{177\text{gs}})\phi t} - N_{177m}(t) \times (\sigma_{\text{burnup}}^m \phi + \lambda_{177m}).$$

The activity of $^{177}\text{Lu}^m$ after an irradiation period was obtained by γ -ray spectroscopy analysis. The obtained averaged burnup cross section value was 568 ± 79 b for the HFR/V4 port experiment. This third value obtained by a very different

method is very close to the burnup values of HFR/T4 and HFR/H9 irradiations. Nevertheless, the HFR/V4 irradiation cannot be compared with the others, because this port is very close to the nuclear fuel and the neutron spectrum is not strictly a Maxwellian flux. This explains the lower measured value in this port since the neutron flux decreases at the lowest energies because of neutron absorption in the fuel.

IV. DISCUSSION

In Tables II and IV, a clear difference exists between the burnup and the radiative capture average cross sections from ORPHEE/p1, HFR/H9 and HFR/T4, HFR/H9 measurements, respectively. A possible contribution to this difference may be the loss of $^{177}\text{Lu}^m$ atoms. This may apply to the nonconfined HFR/H9 target, which has shown no contamination thereby indicating a good stability of the deposit. Since these measurements were performed at thermal energies, only two main processes could explain the disappearance of $^{177}\text{Lu}^m$: radiative capture and INNA reactions.

We will discuss each possible contribution from the burnup, radiative capture, and INNA reactions. We showed in [24] that the radiative capture cross section of the $^{177}\text{Lu}^m$ measured from the activity of $^{178}\text{Lu}^m$ [$J^\pi = (9^-)$] corresponds to the total radiative capture cross section of the $^{177}\text{Lu}^m$. The existence of an unknown long-lived isomer, which could trap a part of the γ decay, is very unlikely because the presence of the $J^\pi = (9^-)$ isomer with a very low energy provides an escape path for any possible high- K spin state. This hypothesis is corroborated by the absence of positive experimental results for new long-lived isomers in ^{178}Lu [31]. Therefore, a probable explanation for the difference between the burnup and the radiative capture cross sections is the INNA reaction. Under this assumption, we deduce the measured value of the INNA reaction cross section by subtracting the values of the capture cross section from the burnup one, for the same neutron flux. The results are presented in the Table V for the two different neutron energy spectra.

Since the deduced cross sections are identical within statistical errors (Table V), the influence of the compound nucleus neutron resonance is negligible. Thus, the resulting cross section for the INNA reaction on $^{177}\text{Lu}^m$ in a Maxwellian flux is a mean value of these two previous measurements

$$\sigma_{\text{INNA}} = 258 \pm 58 \text{ b} \quad \text{at } T = 323 \text{ K.}$$

This measured value is the highest for a inelastic neutron acceleration reaction. The ratio between this INNA cross section and the radiative capture cross section [24] obtained at thermal energy is 0.70 ± 0.17 at $T = 323 \text{ K}$.

TABLE V. Maxwellian averaged INNA reaction cross sections and the ratios between INNA and radiative capture cross sections at 323 K.

Reactor /port	T (K)	f	σ_{INNA} (b)	$\sigma_{\text{INNA}}/\sigma_{(n,\gamma)}$
HFR/T4 – ORPHEE/p1	323	0.0005	254 ± 96	0.69 ± 0.26
HFR/H9 – HFR/H9	323	0.015	261 ± 73	0.71 ± 0.22

At thermal energies, these cross sections are completely determined by the low-lying resonances and are unpredictable. Only statistical distributions around an estimated value could be calculated. Such a work is presented in Ref. [32] where the estimated value for the thermal radiative capture is given by the relation

$$\sigma_{n,\gamma}^* = 0.404 \times 10^8 \left(\frac{A+1}{A} \right)^2 \frac{S_0 \Gamma_\gamma}{D_{\text{CN}}},$$

where A is the mass number of the nucleus, S_0 is the S -wave neutron strength function, Γ_γ is the radiative width, and D_{CN} is the level spacing where CN refers to compound nucleus. The ratio $\Gamma_\gamma/D_{\text{CN}}$ could be obtained from the radiative transmission coefficient by the relation

$$\frac{\Gamma_\gamma}{D_{\text{CN}}} = C \frac{T_\gamma}{2\pi},$$

where C is a normalization constant. The calculations of this transmission coefficient were based on a γ -ray strength function given by Kopecky and Uhl [33]. The C constant was determined after a normalization of the data of the ^{177}Lu resonance parameters [34].

For the INNA reaction, the estimated formula from Ref. [32] is

$$\sigma_{\text{INNA}}^* = 0.404 \times 10^8 \left(\frac{A+1}{A} \right)^2 g(J) S_0^2 \frac{T_{lj}(E_n)}{T_0(E_0)},$$

where $g(J) = (2J+1)/[2(2I+1)]$ is the statistical factor, $T_{lj}(E_n)$ is the neutron transmission coefficient for the neutron energy E_n , and $T_0(E_0)$ is the neutron transmission coefficient at $E_0 = 0.025 \text{ eV}$. We note that in this reference, only the magnetic-type transitions are considered. In $^{177}\text{Lu}^m$, only the magnetic transitions between isomer and final states belonging to the same nucleus are considered.

Using the deformed optical potential developed in Bruyères-le-Châtel [35], calculations were performed to obtain the S -wave neutron strength function $S_0(E)$ and the $T_{lj}(E)$ neutron transmission coefficients. The S -wave neutron strength function was determined for the ^{177}Lu nucleus in the isomer state ($J^\pi 23/2^-$, $S_0 = 1.9886 \times 10^{-4}$) and in the ground state ($J^\pi = 7/2^+$, $S_0 = 1.9912 \times 10^{-4}$). In the case of the isomeric target, the transmission coefficients obtained from the optical model for various orbital momenta carried by the neutron are shown as a function of neutron energy in Fig. 4. For the S -wave neutron, the spin of the compound nucleus formed in the reaction $^{177}\text{Lu}^m(n, n')$ could be either $J^\pi = 11^-$ or $J^\pi = 12^-$. According to Fig. 4, from spin 11^- , the likely neutron transitions correspond to neutron energies of 125.3, 334, and 518.7 keV. From spin 12^- , the neutron transmission coefficients are lower than 10^{-5} .

For these parameters, we obtained the estimated values for the radiative capture and the INNA cross sections, 305 and 27b, respectively. Petrov and Shlyakhter [32] give the probability $S(z)$ that the ratio of the measured cross section to the expected one does not exceed z . In this framework, the probability to

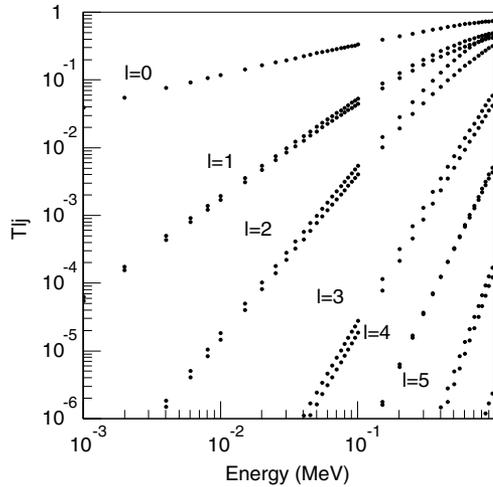


FIG. 4. Neutron transmission coefficients of ^{178}Lu , T_{ij} as a function of neutron energy for various orbital momenta l , at 6.99 MeV, obtained by optical model calculation

obtain the values of cross section higher than the measured radiative capture, 370 b, and INNA cross sections, 258 b, are about 50% and 13%, respectively. According to this statistical approach, one can conclude that the value of the INNA reaction cross section presented in this work on $^{177}\text{Lu}^m$ is plausible. On the other hand, the experimental value of the ratio between the INNA cross section and the radiative capture cross section is high, 0.70, compared to the estimated value, 0.09. As the positions of the resonances are the same for both processes, it means that the overlap between the compound nucleus state

and the final state in ^{177}Lu plays a major role. The observation of the intensity of the various neutron transitions should be a pertinent measurement.

V. CONCLUSION

In conclusion, for the first time, the Maxwellian averaged $^{177}\text{Lu}^m$ burnup cross section, 626 ± 45 b, was measured at the reactor temperature $T = 323$ K. Comparison with the radiative capture cross section allowed us to deduce the cross section of the inelastic neutron acceleration reaction, 258 ± 58 b at $T = 323$ K. It is the highest value ever measured for such a process. We find that the thermal INNA for $^{177}\text{Lu}^m$ is a very efficient way, about 42%, to convert thermal neutrons to high-energy neutrons and thus to deexcite the $^{177}\text{Lu}^m$. This high value of the $^{177}\text{Lu}^m$ INNA cross section provides an opportunity to pursue experiments in order to measure the energy and intensity of the neutron transitions.

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