Thermal neutron capture cross section for the *K* isomer $^{177}Lu^{m}$

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The thermal neutron radiative capture cross section for the *K* isomeric state in ^{177}Lu has been measured for the first time. Several $^{177}Lu^{m}$ targets have been prepared and irradiated in various neutron fluxes at the Laüe Langevin Institute in Grenoble and at the CEA reactors OSIRIS and ORPHEE in Saclay. The method consists of measuring the ^{178}Lu activity by γ-ray spectroscopy. The values obtained in four different neutron spectra have been used to calculate the resonance integral of the radiative capture cross section for $^{177}Lu^{m}$. In addition, an indirect method leads to the determination of the $^{177}Lu^{g}$ neutron radiative capture cross section.

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Nuclear reactions involving isomers are relevant to many topics in nuclear physics [1] such as nuclear reaction mechanisms, nuclear structure, inelastic neutron scattering, astrophysics yields, and nuclear reactor development. When the isomer half-lives are long enough, target preparation is possible and different types of experiments can be performed, for example Coulomb excitation [2], photonuclear reaction [3,4], transfer reactions [5,6], or neutron capture. In particular many long-lived K isomers are known in the $A = 180$ mass region, and these isomers represent a unique chance for energy storage. Their radiative decay rates do not result only from the spin and energy difference with the lower states, but these transitions are hindered by the *K* quantum number difference (*K* being the total spin projection on the nucleus symmetry axis). Hence, we could find processes that are not affected by the *K* hindrance and that could efficiently de-excite a *K* isomer. The thermal neutron superelastic scattering could be such a process, since the *K* number is expected to play no role [7] in the decay of the compound nucleus it goes through. In the neutron superelastic scattering the kinetic energy of the outgoing neutron is greater than that of the ingoing neutron, and the target nucleus has a lower excitation energy in the exit channel. To date, this process has been measured only for ^{152m}Eu [8] and ^{180m}Hf [9].

In the present paper we report measurements of the thermal neutron radiative capture cross section for the ^{177}Lu isomer. This experiment is the first step to obtain the thermal neutron superelastic scattering cross section. The second step will consist of measuring the total burnup cross section in the same neutron spectra. The difference between the cross sections corresponds to the superelastic scattering cross section.

The activation method has been used to measure the capture cross section for the ^{177}Lu isomer (see Fig. 1). Since the ^{178}Lu isomer and ground states half-lives are 28.4 and 23.1 min, the irradiated ^{177m}Lu targets have been γ counted rapidly to measure the total ^{178}Lu activity. After summarizing the target production method, we describe the four different irradiation conditions, and special attention is devoted to the neutron flux measurements and characterizations. We then detail the cross sections measurements in these four neutron spectra. Using these results, we finally extract the neutron radiative capture cross section σ_{γ}^{0} at the neutron velocity of 2200 m s^{-1} and the radiative capture resonance integral I_{γ}.

The ^{177}Lu isomer has been obtained by thermal neutron irradiation of a highly enriched (99.993%) ^{176}Lu powder produced by the mass separator PARSIFAL [10]. Two ^{176}Lu samples with masses 1.25 and 0.31 mg have been irradiated at the ILL high flux reactor (HFR) in Grenoble (France) for 49 days in the V4 irradiation port, where the neutron flux is about 1.5×10^{15} n cm^{-2} s^{-1} with 15% nonthermal neutrons. After irradiation, the samples were cooled to remove the short-lived

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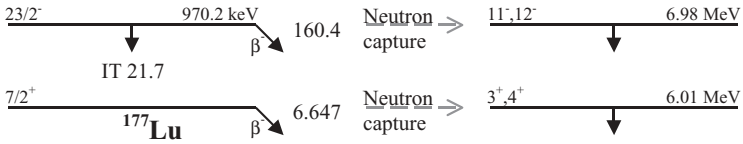
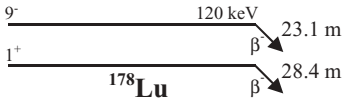


FIG. 1. Scheme of the thermal neutron capture for the ^{177}Lu ground state and isomer.



^{177}Lu ground state (6.647 days) that decays by β emission to ^{177}Hf, which is then chemically removed [11]. Five isomeric targets were produced from these two samples by depositing ^{177}Lu^m on 20 μm thick iron foils. Iron has been chosen because of its low activity after irradiation. Table I summarizes the Five targets' characteristics.

Three tank-type reactors have been used to irradiate these Five targets:

(i) The ORPHEE reactor in Saclay, port p_1: irradiations were limited to 9 min to avoid radiolysis of the plastic container. The maximum attainable flux was about 2×10^{13} n cm^{-2} s^{-1}.

(ii) The OSIRIS reactor in Saclay, ports α_1 and β_1: the main difficulty was to retrieve the samples after a short cooling time. For the two irradiations, the targets were taken out after 94 and 70 min, respectively. Hence the signal/noise ratio was lower on these ports. The fluxes were typically 2×10^{14} n cm^{-2} s^{-1}.

(iii) The ILL high-flux reactor in Grenoble, port H9: this port is equipped with a mechanical carriage that needs 6 min to move the target into the irradiation position and the same time back. Hence the neutron fluence during this time has to be considered. Afterward the targets are automatically placed inside the mini-inca chamber [12] for γ counting. The difficulty was to suppress the γ background due to the 10 g iron target holder: a 25 min irradiation led to a 40 mCi gamma activity. A dedicated collimator was designed to reduce this background because of this huge activity. The neutron fluxes were around 5×10^{14} n cm^{-2} s^{-1}.

Since two ports, α_1 and β_1, have been used on the OSIRIS reactor, four different neutron fluxes were available. Targets 1 and 2 have been irradiated in the same port (ORPHEE p_1). The four irradiation conditions are summarized in Table II. T is the tank water temperature, and f is the epithermal/thermal ratio determined by the k_0 method [13]. For the ILL/H9 irradiation the f ratio has been calculated with a simulated neutron flux. The simulation has been validated by measuring different targets activations (^{59}Co, ^{93}Nb, ^{235}U).

TABLE I. Target characteristics.

Target number	Isomer population (atoms)	Isomer/ground state ratio
1	$(1.71 \pm 0.05) \times 10^{11}$	111.11
2	$(3.05 \pm 0.08) \times 10^{11}$	111.11
3	$(4.5 \pm 0.44) \times 10^{10}$	111.11
4	$(1.22 \pm 0.16) \times 10^{10}$	111.11
5	$(9.40 \pm 0.78) \times 10^{11}$	10

The number of ^{178}Lu nuclei produced by the ^{177}Lu^m (n, γ) reaction has been determined by measuring the ^{178}Hf γ lines coming from the ^{178}Lu β^- decay. In order to get zero dead-time spectra, two different acquisition systems have been used (DSPEC PLUS ORTEC [14] in Saclay, ADONIS [15] in Grenoble). Only γ lines from ^{178}Lu^m have been observed in the spectra. We will see later that the capture level $E_x = 6.98$ MeV, $J^\pi = (11^-, 12^-)$ (see Fig. 1) feeds this isomer only. We will also see that there is no place for another long-lived isomer that could trap a part of the compound nucleus decay. Hence we focus now on the measurement of the ^{178}Lu^m cross section activation, assuming it equals the total radiative capture cross section of ^{177}Lu^m.

The lower part of Fig. 2 shows a fraction of the time-integrated γ-ray spectrum obtained after one of the two ORPHEE p_1 irradiations, with the two γ lines at 325.60 and 327.68 keV assigned to ^{178}Lu^m and ^{177}Lu^m decays, respectively. By use of the ratio between these two γ line areas, the detection efficiency is cancelled out, because the energies are very close. The same elimination is done between the 213.41 (^{178}Lu^m) and 214.43 keV (^{177}Lu^m) γ lines. From the known γ intensities we determine the ratio N_{178m}/N_{177m} of the number of produced ^{178m}Lu nuclei and the number of ^{177m}Lu nuclei present in the target. Then the cross section is calculated according to

$$\sigma_{n,\gamma} = \frac{N_{178m}}{N_{177m}} \times \frac{e^{\lambda_{178} t_{\text{cool}}} (\lambda_{178m} - \lambda_{177m})}{\phi \times e^{t_{\text{irr}} \cdot \lambda_{177m}} (e^{-\lambda_{177m} t_{\text{irr}}} - e^{-\lambda_{178m} t_{\text{irr}}})(1 - e^{-\lambda_{178m} t_{\text{count}}})},$$

where t_{irr} is the irradiation time, t_{cool} is the cooling time, and t_{count} is the counting time. By neglecting the ^{177}Lu^m decay during the measurement, we can write

$$\sigma_{n,\gamma} = \frac{N_{178m}}{N_{177m}} \frac{\lambda_{178m} e^{\lambda_{178} t_{\text{cool}}}}{\phi (1 - e^{-\lambda_{178m} \times t_{\text{irr}}})(1 - e^{-\lambda_{178m} t_{\text{count}}})}.$$

TABLE II. Characteristics of the different experiments at ORPHEE, OSIRIS, and ILL reactors. The first row indicates that targets 1 and 2 were both irradiated in the ORPHEE p_1 port but with different cooling times.

Target no.	Reactor port	T (K)	Flux monitor	Total flux (n cm^{-2} s^{-1})	f	Irradiation time (min)	Cooling time (min)
1/2	ORPHEE p_1	323	IRMM AlAu	1.077×10^{13}	0.0005	9	6/13
3	OSIRIS β_1	308	Fe and Zr foils	1.37×10^{14}	0.031	26	94
4	OSIRIS α_1	308	Fe and Zr foils	8.62×10^{13}	0.029	26	70
5	ILL H9	323	IRMM AlCo	6.23×10^{14}	0.015	23	21.3

Half-life values of 23.1 ± 0.3 min for the $^{178}Lu^m$ and 160.44 ± 0.06 days for the $^{177}Lu^m$ have been used, as published in ENSDF [16].

In order to prove that these γ lines are not contaminated, three techniques have been used. The first one consists of verifying that the time dependency of the 214.41 and 325.6 keV γ lines corresponds to the $^{178}Lu^m$ half-life. The second one consists of checking the consistency of all the $^{178}Lu^m$ γ-ray lines (88.85, 93.15, 213.41, and 325.6 keV) according to the known intensities. The last technique consists of assigning all the γ lines of the spectra to a known nucleus and verifying that these nuclei do not have any γ lines close to 214.41 and 325.6 keV.

Figs. 2 and 3 show the 325 keV γ-line decay and the γ spectrum around the 325-327 keV doublet obtained for the ORPHEE p_1 and ILL H9 irradiations, respectively. The two fitted half-lives (22.7 ± 1.4 min, and 23.7 ± 1.7 min, respectively) are consistent with the ENSDF value of 23.1 ± 0.3 min.

Fig. 4 shows the γ-ray spectrum near the 325-327 keV doublet for the OSIRIS irradiation. In that case, owing to a lower signal, no half-life value could be extracted. Only the 213.41 and 325.6 keV lines could be used to check the intensities according to the known branching ratios.

For each irradiation, the signal/background ratio is different, depending on the neutron fluences and cooling time combinations. Table III summarizes the measured effective radiative capture cross sections for $^{178}Lu^m$ activation for each reactor port. The given cross sections are weighted averages of the cross sections obtained from the 213-214 and 325-327 keV doublets.

The ORPHEE p_1 value has been calculated with the two measurements for targets 1 and 2. For ILL H9, a 0.98 factor has been calculated to correct for the neutron fluence during the sample transfer in-out and to-from the irradiation position. This correction is small compared with the uncertainties.

These effective cross sections, measured in several types of neutron flux, have been used to determine the neutron radiative

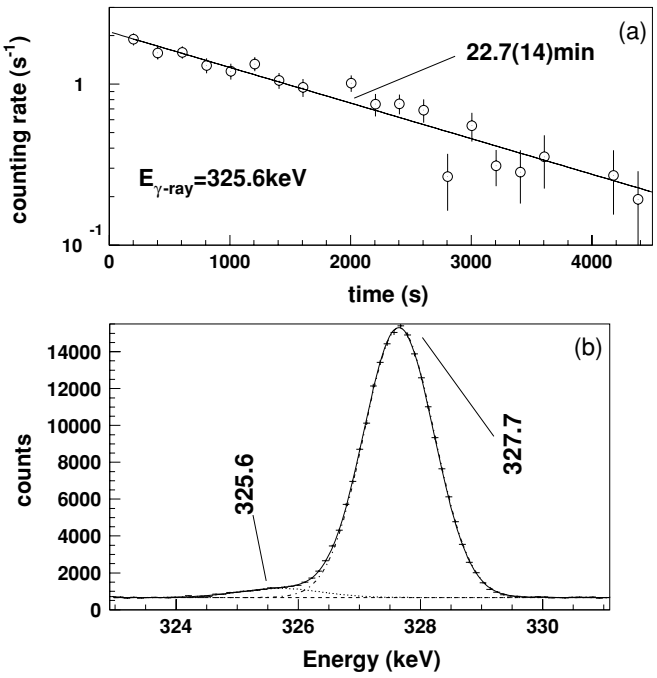


FIG. 2. ORPHEE p_1 irradiation. (a) Fitted half-life on the 325 keV γ-line intensity as a function of time. (b) Partial γ-ray spectrum obtained for the irradiated target. The two γ-lines at 325.60 and 327.68 keV are from the $^{178}Lu^m$ and $^{177}Lu^m$ decays, respectively.

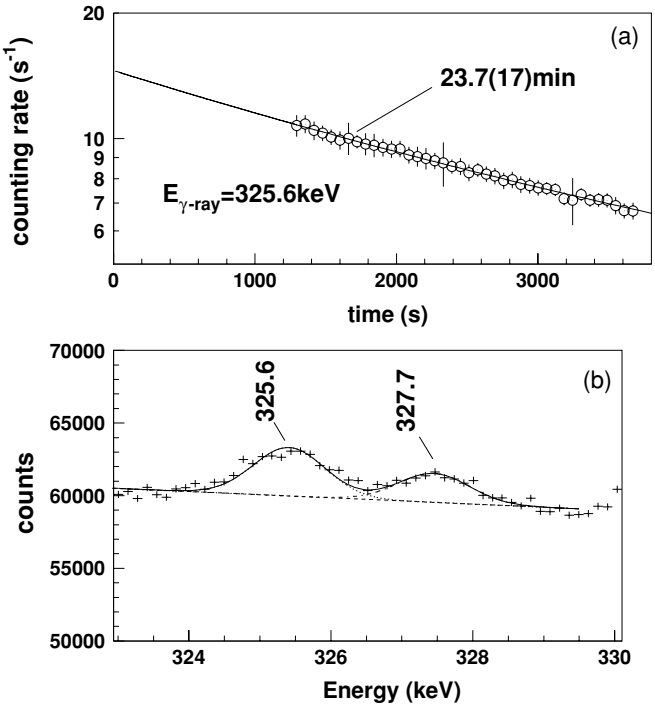


FIG. 3. ILL H9 irradiation. (a) Fitted half-life of the 325 keV γ-line intensity as a function of time. (b) Partial γ-ray spectrum obtained for the irradiated target.

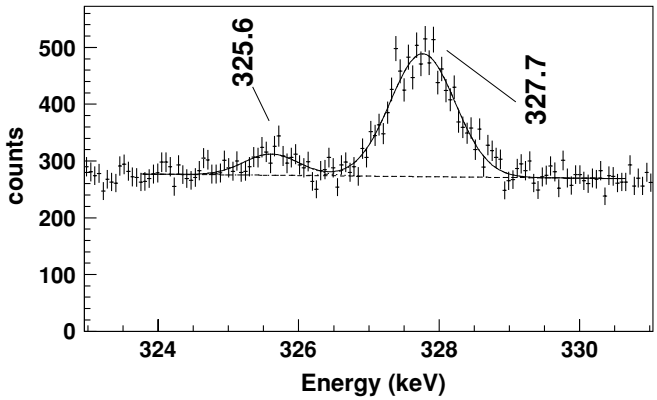


FIG. 4. OSIRIS α_1 irradiation: partial γ-ray spectrum.

capture cross section σ_γ^0 at the neutron velocity of 2200 m s^{-1} and the radiative capture resonance integral I_γ. In Westcott's convention [17] the effective cross section is

$$\hat{\sigma}_{n,\gamma} = \sigma_\gamma^0 \left(g_w + r\sqrt{\frac{4T}{\pi T_0}}\frac{I'_\gamma}{\sigma^0} \right),$$

where g_w is the Westcott factor, $r = f\sqrt{\pi\mu/4}$, $T_0 = 293.6$ K, and I'_γ is the reduced resonance integral. The variable μ is an energy cutoff parameter that defines the epithermal neutron domain. Its value, which has been set at 5 eV [17], has a very little influence on the final result for the capture cross section.

From Table III we see that the temperatures in the ILL and ORPHEE reactors are the same, so the temperature-dependent Westcott factors are the same. Since the epithermal/thermal ratio is not the same, these two measurements provide a coupled equation system where $\sigma_0 g_w$ and I'_γ are the unknown variables. The solution is

$$\begin{aligned} \sigma^0 g_w^{323K} &= 418 \pm 29 \text{ barns}, \\ I'_\gamma &= -300 \pm 3900 \text{ barns}. \end{aligned} \quad (1)$$

Since I'_γ must be positive, it will be considered equal to zero in the following analysis. Hence the two OSIRIS irradiation results (Table III) lead to

$$\sigma^0 g_w^{308K} = 413 \pm 58 \text{ barns}. \quad (2)$$

From these four measurements we conclude that the compound nucleus resonances have a very little effect on the neutron radiative capture for ^{177}Lu^m: the resonance integral is very low, and the Westcott factor does not vary significantly between the two temperatures 308 and 323 K. If we assume that $g_W = 1$, we obtain, by averaging results (1) and (2) for the two different temperatures,

$$\sigma_\gamma^0 = 417 \pm 26 \text{ barns}.$$

This cross section for the ^{177}Lu^m (n,γ) reaction has been obtained from the ^{178}Lu^m activity. This result is valid if there is no significant ^{178}Lu^g production in the ^{177}Lu^m irradiation and there is no possible unseen γ decay after neutron capture. A γ-ray cascade code has been used to follow the γ-decay path from the capture level (excitation energy of 6.98 MeV and $J^\pi = (11^-, 12^-$, see Fig. 1) down to the known low energy levels [18]. A very high probability, 99.7%, has been obtained for the $K^\pi = 9^-$ isomer in ^{178}Lu feeding. Moreover, the existence of an unknown long-lived isomer that could trap a part of the γ decays is very improbable because of the presence of the $K^\pi = 9^-$ isomer with a very low energy that provides an escape path for any possible high-K state. Any other isomer could decay to a rotational state built on this isomer, with low forbideness factors $\nu = \Delta K - \lambda$, where ΔK and λ are the transition K variation and multipolarity. Therefore such a state should be short lived and should have been seen in our spectra. This hypothesis is supported by the negative result of a recent experiment [19] searching for new isomers in that nucleus.

Moreover, the ^{178}Lu^m activity could be affected by the neutron capture on ^{177}Lu^g, since the targets contain a small amount of these nuclei. The cross section of this process has been indirectly measured by using the ^{176}Lu irradiation. The initial ^{176}Lu activity has been measured before irradiation, and the ^{177}Lu^g activity measured after irradiation. with this known isotope production cross section and half-life, a value of 880 $\pm$ 75 barns has been obtained, which is compatible and much more precise than the published value of 1000 $\pm$ 300 barns [20]. Again, the γ-ray cascade code has been used to deduce the ^{178}Lu^m activity due to the neutron capture by the ^{177}Lu ground state. In the target having the largest ^{177}Lu^g population, this activity represents 3.44% of the ^{178}Lu^m activity. This value is far less than the ^{177}Lu^m (n,γ) cross section uncertainties, and no correction has been made to take this effect into account.

The presented work represents the first measurement of the neutron radiative capture on the ^{177}Lu^m state, for which a value of 417 $\pm$ 26 barns has been obtained. The ^{177}Lu^g $(n,\gamma)^{178}$Lu has been determined as 880 $\pm$ 75 barns. This work is the first step to measure the superelastic scattering cross section for ^{177}Lu^m. The next step will consist of measuring the burnup cross section in similar neutron fluxes to obtain an indirect measurement of that cross section by difference.

TABLE III. Summary of the effective radiative capture cross sections measurements on ^{177}Lu^m.

Reactor port	T (K)	Neutron flux (n cm^{-2} s^{-1})	f	$\hat{\sigma}_{n,\gamma}$ (barns)
ORPHEE p1	323	1.077 10^{13}	0.0005	418 $\pm$ 28
OSIRIS β1	308	1.16 10^{14}	0.031	411 $\pm$ 97
OSIRIS α1	308	7.21 10^{13}	0.029	424 $\pm$ 73
ILL H9	323	5.52 10^{14}	0.015	413 $\pm$ 60

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