EPJ Web of Conferences **146**, 11048 (2017) DOI: 10.1051/epjconf/201714611048
ND2016

Cross section of the ^{197}Au(n,2n)^{196}Au reaction

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Abstract. The ^{197}Au(n,2n)^{196}Au reaction cross section has been measured at two energies, namely at 17.1 MeV and 20.9 MeV, by means of the activation technique, relative to the ^{27}Al(n,α)^{24}Na reference reaction cross section. Quasi-monoenergetic neutron beams were produced at the 5.5 MV Tandem T11/25 accelerator laboratory of NCSR "Demokritos", by means of the ^{3}H(d,n)^{4}He reaction, implementing a new Ti-tritiated target of ∼400 GBq activity. The induced γ-ray activity at the targets and reference foils has been measured with HPGe detectors. The cross section for the population of the second isomeric (12^-) state m2 of ^{196}Au was independently determined. Auxiliary Monte Carlo simulations were performed using the MCNP code. The present results are in agreement with previous experimental data and with theoretical calculations of the measured reaction cross sections, which were carried out with the use of the EMPIRE code.

1. Introduction

Accurate neutron-induced reaction cross section data is of considerable importance for fundamental research in Nuclear Physics and Astrophysics as well as for practical applications in nuclear technology, medicine and industry [1,2]. Neutron induced reactions on Au, especially (n,2n), are proposed as a standard for high energy neutron dosimetry [3] and are also very important as reference-monitor reactions. Moreover, the presence of a high-spin (12^-) isomeric state (m2) in the residual nucleus of the ^{197}Au(n,2n) reaction, makes it a sensitive test-case for several nuclear reaction model codes [4,5].

Although a lot of cross section data exist in the energy region up to 14 MeV, more experimental data is needed, especially for the second isomeric state (m2) at higher energies, above 15 MeV, where the pre-equilibrium emission becomes important and additional reaction channels are open.

Thus, the purpose of this work was the experimental determination of the cross section of the reaction channels ^{197}Au(n,2n)^{196}Au and ^{197}Au(n,2n)^{196}Au^{m2} at incident neutron energies 17.1 and 20.9 MeV, implementing the activation technique. Preliminary results from the 17.1 MeV measurement along with the neutron beam characterization at this energy, have been presented in Ref. [6]. The cross section for the population of the second isomeric (m2) state of ^{196}Au was independently determined. The neutron beam was produced at the 5.5 MV tandem T11/25 Accelerator of NCSR "Demokritos" by means of the ^{3}H(d,n)^{4}He reaction, implementing a new Ti-tritiated target of ∼400 GBq activity. Furthermore, theoretical statistical model calculations were performed and compared to all available experimental data in literature.

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2. The reaction

When a neutron with an energy 17.1 or 20.9 MeV impinges on a ^{197}Au nucleus, the compound nucleus ^{198}Au is produced in a highly excited state and the possible exit channels of the reaction are shown in Fig. 1.

In the present work the ^{197}Au(n,2n)^{196}Au channel was studied, in which the residual nucleus is produced in an excited state and presents an interesting isomeric pair, that is ground and isomeric states (g, m1, m2) with spin values of 2^-, 5^+ and 12^-, respectively. The ground state (g) decays to ^{196}Pt with a half life of 6.2 d and its population is deduced through the 355.7 keV transition, prefered over the 333.0 keV gamma ray, due to its high intensity. The first isomeric state (m1) at 86.7 keV excitation energy has a short half life 8.1 s, thus its population cannot be measured separately from the population of the ground state. The second isomeric state (m2), at 595.7 keV excitation energy, decays to the ground state via several transitions with the 147.8 keV and 188.3 keV being the most intense ones and are used for the determination of its population by the ^{197}Au(n,2n) reaction.

3. Activation and mesurements

The ^{197}Au(n,2n)^{196}Au reaction cross section has been measured at 17.1 and 20.9 MeV, by means of the activation technique, relative to the ^{27}Al(n,α)^{24}Na reaction reference cross section, at the 5.5 MV Van de Graaff Tandem T11/25 accelerator laboratory of NCSR "Demokritos".

Quasi-monoenergetic neutrons were produced by means of the ^{3}H(d,n)^{4}He reaction, implementing a new Ti-tritiated target consisting of a 2.3 mg/cm^2 Ti-T layer on a 1 mm thick Cu backing for good heat conduction. The flange with the tritium target assembly was air-cooled during the irradiations and the Au foil was stacked between two Al foils and placed at a distance of ∼2 cm from the

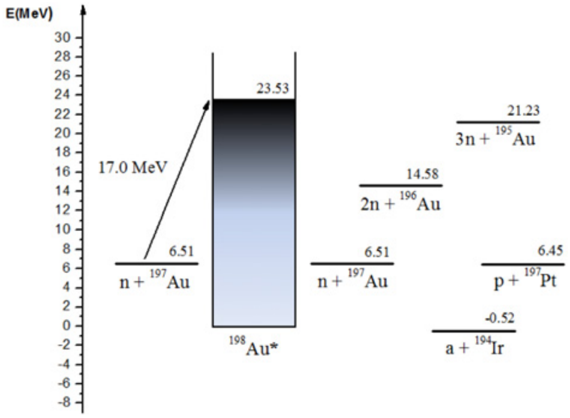


Figure 1. Energy diagram of the n+^{197}Au interaction.

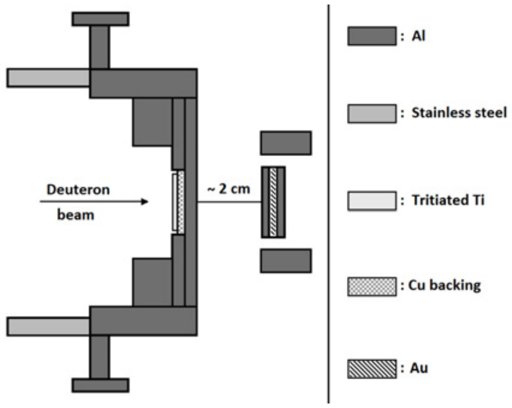


Figure 2. Schematic representation of the irradiation setup geometry.

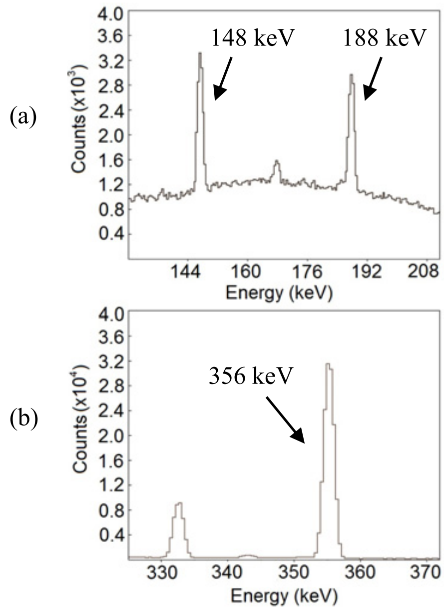


Figure 3. Typical off-line spectra of natural Au taken at incident neutron energy 17.1 MeV, where the gamma-ray peaks from the decay of the second isomeric state (a) and both ground and isomeric states (b) of ^{196}Au nucleus are presented. The acquisition time is 18.0 and 25.0 h, respectively.

Table 1. Decay properties of the daughter nuclei [7,8].

Daughter nucleus	Half-life	γ-ray energy (keV)	Intesity per decay (%)
^{196}Au	6.2 d	355.7	87.0
^{196}Au ^{m2}	9.6 h	147.8	43.5
		188.3	30.0
^{24}Na	15.0 h	1368.6	100.0

tritium target (Fig. 2). All samples were high purity foils of ∼0.5 mm thickness and ∼1.2 cm diameter.

In this way, the angular acceptance of the target and reference foils was limited to ±15°, therefore the produced neutrons are expected to be practically monoenergetic according to the reaction kinematics. The beam current on the targets was 1 μA and 300 nA, resulting in a neutron flux of 3×10^5 and $2 \times 10^5 \mathrm{n/cm^2\,s}$ for the irradiations at 17.1 and 20.9 MeV, respectively. The former irradiation lasted for 96.1 h, while the latter for 32.4 h, during which the neutron beam was monitored by a BF_3 detector, placed at a distance of 3 m from the neutron source. The output of the BF_3 detector was stored at regular time intervals and was used to correct for the decay of ^{196}Au nuclei during the irradiations and to account for fluctuations in the beam flux in the off-line analysis.

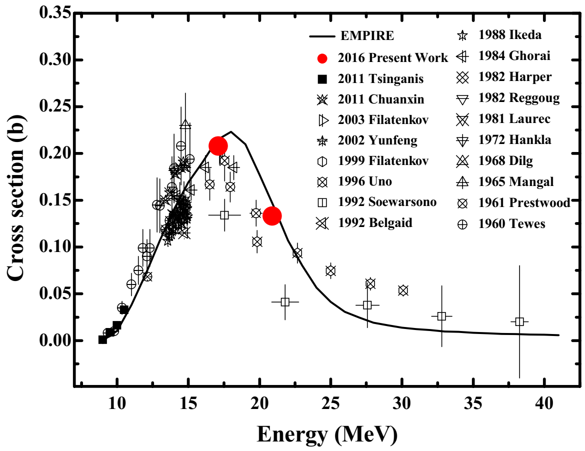
After the irradiations, the induced activity on aluminum and gold samples was measured with two HPGe detectors of 100% and 56% relative efficiency, properly shielded with lead blocks to reduce the contribution of the natural radioactivity. The samples were placed at a distance of 10 cm from the detector window and a calibrated ^{152}Eu source, placed at the same distance, was used to determine the efficiency of the detectors. With this setup, corrections for true coincidence summing effects were negligible. Typical spectra of the gold samples are shown in Fig. 3(a) and (b), where the gamma-rays of interest have been marked. The gamma- ray intensities and half-lives used in the analysis are presented in Table 1. The measurements for the second isomeric state (m2) started ∼1 h after the end of the irradiation and the corresponding cross sections were derived via the 147.8 and 188.3 keV gamma-ray peaks. Due to the fact that the first isomeric state (m1, $T_{1/2} = 8.1$ s) decays with a relatively short half-life, the measurement of the decay of the ground state (g) contains the population of both the ground and the first isomeric states (g + m1). Moreover, since the measurements for the ground and the first isomeric state (g + m1) began ∼40 h after the irradiation, so that the second isomeric state (m2, $T_{1/2} = 9.6$ h) had fully decayed to the ground state (g), the population of all of these states: ground, first and second isomeric (g + m1 + m2) was evidently included in the final result, i.e., the cross section which was obtained through the analysis of the 355.7 gamma- ray peak. Additional corrections for the self-absorption inside the samples were taken into account by using the MCNP5 code [9].

4. Theoretical calculations and results

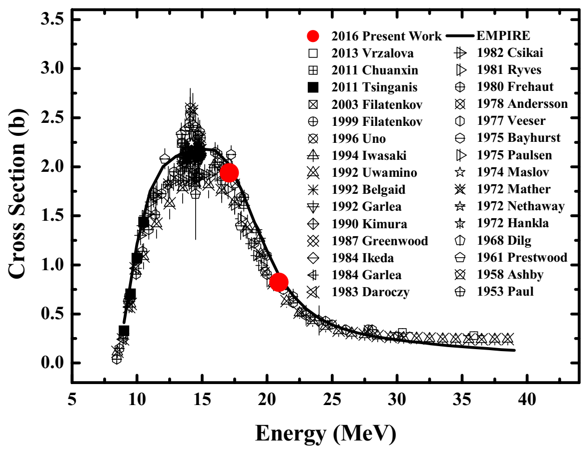
Theoretical cross section calculations in the energy range between 7 and 50 MeV were performed by means of the EMPIRE code (version 3.2.2, "Malta") [18].

Compound nucleus reaction calculations were performed in the framework of the Hauser-Feshbach theory [10]. The EMPIRE-specific level densities were implemented for the description of the continuous excitation spectra of the nuclei at equilibrium deformation, while

DOI: 10.1051/epjconf/201714611048



(a)



(b)

Figure 4. Experimental values of the present work at 17.1 and 20.9 MeV incident neutron energy, along with EXFOR data [19] from literature and theoretical calculations for: (a) the population of the second isomeric state of ^{196}Au (m2), (b) the population of the ground, first and second isomeric states of ^{196}Au.

the correlation between the incident and exit channels in elastic scattering (width fluctuation corrections) was also taken into account according to the HRTW model [11]. The optical model parameters for outgoing particles were taken from the Reference Input Parameter Library (RIPL-3) database [12] using the data by D. Wilmore et al. [13], A.J. Koning et al. [14] and V. Avrigeanu et al. [15], for outgoing neutrons, protons and alphas, respectively, while gamma-ray strength functions were also used. For the outgoing neutrons, eighteen potentials, available in RIPL-3 have been tested and the most suitable OMP for this application, apart from the chosen one (D. Wilmore et al.), was one suggested by O. Bersillon et al. [16], while the local OMP by A.J. Koning et al. overestimated the cross section of the second isomeric state.

Concerning the direct reaction mechanism, the population of discrete collective levels for inelastic scattering was described by the coupled-channel method and was calculated using the ECIS03 code [17]. The default option was used, which provides spherical optical model parameters and suppresses the calculation of direct reactions to the discrete levels.

For the pre-equilibrium contribution, although both the quantum-mechanical formulations (MSD and MSC) were tried in several different combinations simultaneously with the OMP tests, the most appropriate choice for our case was the classical approach, by means of the exciton model as implemented in the EMPIRE code through the PCROSS module [18].

It should be emphasized that the ground state (g + m1 + m2) cross section data could be well reproduced by many combinations of optical and level density models, while the isomeric (m2) cross section data could not be easily described by the theoretical calculations. Eventually, by using the aforementioned parametrization, the results of the EMPIRE code reproduced fairly well, not only the existing experimental data in (n,2n) channels (Fig. 4(a), 4(b)), but also three additional reaction channels, namely the: (n,3n), (n,p) and (n,α).

The preliminary experimental results for the cross section of the ^{197}Au(n,2n)^{196}Au^{m2} and total ^{197}Au(n,2n)^{196}Au reactions, are presented in Fig. 4(a) and Fig. 4(b) respectively, along with EXFOR data from literature [19] and the theoretical calculations which were performed with the EMPIRE code.

The experimental uncertainties of all the factors used in the determination of the cross section errors, are summarized in Table 2 and were summed quadratically.

Table 2. Compilation of uncertainties.

	Uncertainty (%)
Neutron energy	< 1
Neutron integrated flux[1]	4–5
Counting statistics for total (n,2n) γ-ray peaks	< 1
Counting statistics for m2 γ-ray peaks	< 2
Detector efficiency	3
Total uncertainty of cross section for total (n,2n)	4–6
Total uncertainty of cross section for m2	5–6

[1] Including 3% uncertainty in the ^{27}Al(n,a)^{24}Na cross section.

5. Summary

The cross section of the (n,2n) reaction on ^{197}Au was measured independently for the population of the second isomeric state (m2) and for the sum of the reaction cross sections for the population of ground, first and second isomeric states. The new measurements performed by means of the activation technique, relative to the ^{27}Al(n,α)^{24}Na reference reaction cross section. Quasi-monoenergetic neutron beams were produced at the 5.5 MV Tandem T11/25 accelerator laboratory of NCSR “Demokritos” at incident neutron energies of 17.1 and 20.9 MeV. The preliminary experimental results are presented in Fig. 4(a) and Fig. 4(b) and follow the general trend of the existing experimental data. Furthermore, theoretical calculations in the energy range between 7 and 50 MeV were performed by means of the EMPIRE code for the (n,2n) channel, which successfully reproduced the (n,3n), as well as the (n,p) and (n,α) channels. More measurements are planned at ∼15 and 19 MeV neutron energies, which will cover the high energy range and will help to resolve discrepancies among the existing experimental data.

Special thanks to the accelerator staff at NCSR "Demokritos" for the invaluable assistance.

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