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EXPERIMENTAL STUDY AND THEORETICAL CONSIDERATION OF THE ISOMERIC RATIO IN PHOTONUCLEAR REACTION ^{197}Au$(\gamma, n)^{196m,g}$Au IN THE GIANT DIPOLE RESONANCE REGION

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We have carried out the study on isomeric ratio in ^{197}Au$(\gamma, n)^{196m,g}$Au photonuclear reaction in the giant dipole resonance region with bremsstrahlungs produced by electron accelerator Microtron MT-25 of the Flerov Laboratory of Nuclear Reactions, JINR. The isomeric ratio has been determined from a series of gamma spectra measured with high energy resolution gamma spectrometric system consisting of HPGe detector and a multichannel analyzer. The results have been discussed, compared with those of other authors as well as considered by the statistical model proposed by Huizenga and Vandenbosh.

Проведено исследование изомерного отношения в фотоядерной реакции ^{197}Au$(\gamma, n)^{196m,g}$Au в области гигантского дипольного резонанса. Работу проводили на микротроне МТ-25 ЛЯР ОИЯИ. Изомерное отношение определяли из серии гамма-спектров, измеренных HPGe-детектором с многоканальным анализатором. Проведено сравнение полученных данных с результатами других авторов и статистической моделью Хьюзенги и Ванденбоша.

INTRODUCTION

In (γ, n) photonuclear reaction in the giant electric dipole resonance (GDR) region by interaction with photon flux, the absorption of an $E1$ γ-ray brings a target nucleus with spin J_0 to excited state in the first compound nucleus A^* with spin $J_c = J_0$, $J_0 \pm 1$. For $J_0 = 0$, only states with $J_c = \pm 1$ are excited. The spin distribution in the first compound system A^* is changed strongly by the successive evaporation of one or more neutrons, and the product nucleus is formed. Usually the product nucleus with an isomeric state and an unstable ground state may be used to measure the relative population of these two states in nuclear reaction, the so-called isomeric cross-section ratio.

The isomeric ratios furnish valuable information about the energy level structure of nuclei and the nuclear reaction mechanism involved. On the other hand, the isomeric and unstable ground states are formed simultaneously during nuclear reaction process in the same experimental conditions, so the isomeric ratios can be determined with high accuracy. By fitting the calculated isomeric ratios to the experimental ones, it is possible to obtain information about the spin dependence of the nuclear level density, in particular, the spin cut-off parameter σ and the level density parameter a.

In the previous works [1–5], we have investigated isomeric ratios in different nuclear reactions and considered the results by the statistical model proposed by Huizenga and Vandenbosh [6, 7]. These investigations led to very interesting conclusions about angular momentum effect, nuclear reaction mechanism and correlation between spin cut-off parameter σ and centre of spins (COS).

The purpose of this work is to study the isomeric ratio in $^{197}\mathrm{Au}(\gamma, n)^{196m,g}\mathrm{Au}$ photonuclear reaction in the giant dipole resonance region. In principle the relative population of the isomeric state and unstable ground state strongly depends on the spin difference of these states. In case of $^{196m,g}\mathrm{Au}$ this difference is very high; therefore, it is expected that in the GDR region the isomeric ratio is very low. Besides we have also considered our results by the method of Huizenga and Vandenbosh.

1. EXPERIMENTS

Our experiments were carried out using the bremsstrahlungs produced by electron accelerator Microtron MT-25 of the Flerov Laboratory of Nuclear Reaction, Joint Institute for Nuclear Research, Dubna, Russia. The bremsstrahlung end-point energy can be varied stepwise from 10 to 25 MeV. The description of this accelerator and its main characteristics are presented in [8]. The essential advantage of this Microtron is the small energy spread of the accelerated electrons (30–40 keV) at high beam intensity (up to an average power of 600 W). This allows measurement of the isomeric ratio of the studied nuclides production at strictly definite end-point energy. As an electron–photon converter was used W disk 4 mm in thickness, cooled by water. For absorption of electrons passing in the reaction cameras, an aluminum screen 20 mm in thickness was placed behind the converter.

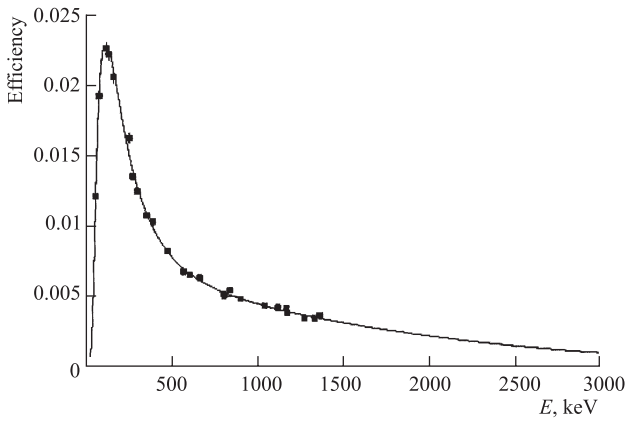

Fig. 1. The efficiency of HPGe detector CANBERRA measured at a distance of 5 cm

The investigated samples were prepared of 99.99%-purity gold foils in disk form 1 cm in diameter. In order to study isomeric ratio in the GDR region, four gold samples were irradiated with bremsstrahlung of end-point energies 15, 18, 21.5, and 23.5 MeV, and the average accelerated electron beam was about 15, 10, and 12 μA respectively. The irradiation time for the above-mentioned bremsstrahlungs was 40, 30, 10, and 20 min respectively.

The gamma spectra of the reaction product were measured with a spectroscopic system consisting of HPGe detector CANBERRA with energy resolution of 1.8 keV at 1332-keV gamma ray of $^{60}\mathrm{Co}$, amplifier 2022 and multichannel analyzer 8192 connected to computer for data

processing. The efficiency of the detector was determined by using a set of single gamma-ray sources calibrated to 1–2% and is shown in Fig. 1.

Table 1. **The characteristics of the product of ^{197}Au$(\gamma, n)^{196m,g}$Au photonuclear reaction**

Reaction	Product	Half-life	Spin $\hbar$	Reaction threshold, MeV	Energy, keV	Intensity, %
^{197}Au(γ, n)	^{196m}Au	9.7 h	12^-	8.68	147.7 168.3 188.2	43.00 7.74 37.84
^{197}Au(γ, n)	^{196g}Au	6.183 d	2^-	8.09	333.0 355.0 426.0	27.15 87.60 6.75

The characteristics of the products of ^{197}Au$(\gamma, n)^{196m,g}$Au are taken from [9, 10] and presented in Table 1.

2. ISOMERIC RATIO CALCULATION

The process of producing the nuclei in isomeric state and unstable ground state in a nuclear reaction can be described by the following equations:

$$\begin{aligned} \frac{dN_m}{dt} &= N_0\phi\sigma_m - \lambda_m N_m, \\ \frac{dN_g}{dt} &= N_0\phi\sigma_g - \lambda_g N_g + P\lambda_m N_m, \end{aligned} \tag{1}$$

where N_0 is the number of the nuclei concerned in target; ϕ is the flux of the projectile particles; N_m, N_g are the numbers of nuclei in the isomeric state and in the unstable ground state; λ_m, λ_g are their decay constants respectively; P is the isomeric transition coefficient.

Resolving the above equations for definite irradiation, cooling and measurement times, we get an analytical expression for the isomeric ratio as follows:

$$\frac{1}{R} = \frac{\dfrac{S_g\varepsilon_m I_m}{S_m\varepsilon_g I_g}\Lambda_3\Lambda_6\Lambda_9 - \Lambda_1\Lambda_5\Lambda_8 - \Lambda_3\Lambda_5\Lambda_8 - \Lambda_3\Lambda_6\Lambda_7}{\Lambda_2\Lambda_5\Lambda_8}. \tag{2}$$

Here m and g are isomeric and unstable ground states; I, ε and S are the intensity, the efficiency and the area under the photopeak concerned respectively; R is the isomeric ratio. And $\Lambda_1, \ldots, \Lambda_9$ are the factors connected to the times of irradiation, cooling and measurement and are expressed as

$$\Lambda_3 = \frac{1}{\lambda_m}\left(1 - \mathrm{e}^{-\pi_m t_1}\right),$$

$$\Lambda_6 = \mathrm{e}^{-\lambda_m t_2},$$

$$\Lambda_9 = 1 - \mathrm{e}^{-\lambda_m t_3},$$

$$\Lambda_1 = \frac{P}{\lambda_g}\left[\left(1 - \mathrm{e}^{-\lambda_g t_1}\right) - \frac{\lambda_g}{\lambda_m - \lambda_g}\left(\mathrm{e}^{-\lambda_g t_1} - \mathrm{e}^{-\lambda_m t_1}\right)\right],$$

$$\Lambda_2 = \frac{1}{\lambda_g}\left(1 - \mathrm{e}^{-\lambda_g t_1}\right),$$

$$\Lambda_4 = \frac{P\lambda_m}{\lambda_m - \lambda_g}\left(\mathrm{e}^{-\lambda_g t_2} - \mathrm{e}^{-\lambda_m t_2}\right),$$

$$\Lambda_5 = \mathrm{e}^{-\lambda_g t_2},$$

$$\Lambda_7 = P\left[1 - \frac{\lambda_m \lambda_g}{\lambda_m - \lambda_g}\left(\frac{\mathrm{e}^{-\lambda_g t_3}}{\lambda_g} - \frac{\mathrm{e}^{-\lambda_m t_3}}{\lambda_m}\right)\right],$$

$$\Lambda_8 = \left(1 - \mathrm{e}^{-\lambda_g t_3}\right).$$

Here t_1, t_2, t_3 are the irradiation, cooling and measurement times.

In calculation, for simplified case we suppose that in the experiment the irradiation, cooling and measurement times are the same for the isomeric and metastable states.

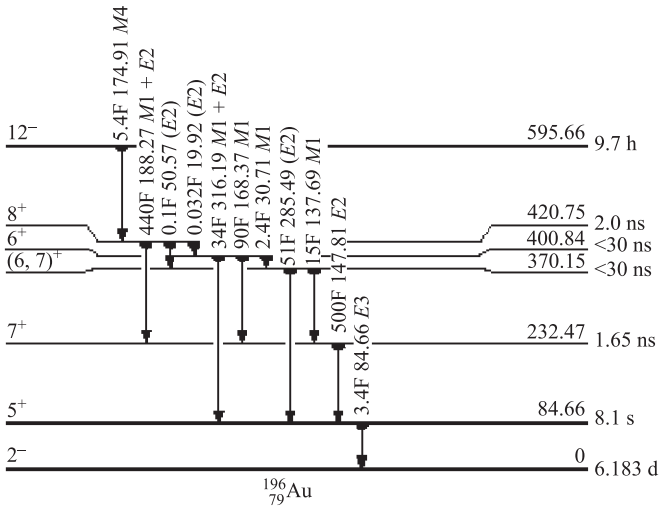

Fig. 2. Decay scheme of the isomeric pair ^{196m,g}Au

In practice, when there exists a big difference in the half-lives of the isomeric and unstable ground states, the irradiation, cooling and measurement times will be different for these states depending on their half-lives. In this case we have Λ_i^m and Λ_i^g for the isomeric and unstable ground states respectively and the isomeric ratio has the following expression:

$$\frac{1}{R} = \frac{\dfrac{S_g \varepsilon_m I_m}{S_m \varepsilon_g I_g}\Lambda_3^m \Lambda_6^m \Lambda_9^m - \Lambda_1^g \Lambda_5^g \Lambda_8^g - \Lambda_3^g \Lambda_5^g \Lambda_8^g - \Lambda_3^g \Lambda_6^g \Lambda_7^g}{\Lambda_2^g \Lambda_5^g \Lambda_8^g}. \tag{3}$$

Putting the measured gamma spectra data and the data from Table 1 into (3), for the isomeric and unstable ground states one can determine the isomeric ratio.

Figure 2 shows the decay scheme of the isomeric pair ^{196m,g}Au, taken from [9].

3. THEORETICAL MODEL CONSIDERATION

Though the statistical model proposed by Huizenga and Vandenbosh [6, 7] was established long ago, it is still very powerful and widely applied tool for nuclear reaction investigations [17, 18].

In the Huizenga and Vandenbosh model, the theoretical isomeric ratios are calculated as follows:

$$R = \frac{\sum\limits_{J_f=\mathrm{COS}}^{\infty} P\left(J_f\right)}{\sum\limits_{J_f=\mathrm{COS}}^{M} P\left(J_f\right)}, \tag{4}$$

where COS $= (S_h + S_l)/2$, S_h and S_l are spins of high and low spin states of isomeric pair; $P(J)$ is the probability for forming an ensemble of states with different final spin J after the successive evaporation of one neutron from compound nucleus.

We begin by considering the following steps for special case of (γ, n) photonuclear reaction:

• Absorption of an $E1$ γ-ray brings a target nucleus with spin J_0 to forming an excited compound nucleus with the relative occupation probability:

$$P(J_c) \approx 2J_c + 1, \quad J_c = J_0, J_0 - 1, J_0 + 1. \tag{5}$$

• Relative probability for forming a residual nucleus with spin J after the successive evaporation of one neutron is

$$P\left(J\right) \approx \rho\left(J, E^*\right) \sum_{J_c} P\left(J_c\right) \sum_{S=|J-1/2|}^{J+1/2} \sum_{J_c} P\left(J_c\right) \sum_{l=|S-J_c|}^{S+J_c} T_l\left(E_n\right), \tag{6}$$

where $T_l(E_n)$ is the penetrability for a neutron with angular momentum l and kinetic energy E_n; $\rho(E^*, J)$ is the level density of excited nucleus, represented by the following formula:

$$\rho\left[E^*, J\right] = \rho\left(E^*\right)\rho\left(J\right) \sim \exp\left[2\left(aE^*\right)^{1/2}\right]\left(2J+1\right)\exp\left[-\frac{J\left(J+1\right)}{2\sigma^2}\right], \tag{7}$$

where E^* is the excitation energy of residual nucleus; σ is the spin cut-off parameter and a is the level density parameter.

In calculation, we supposed that the energy distribution of the emitted neutrons has Maxwell form and the average kinetic energy is

$$\overline{E_n} = \frac{1 + \sqrt{1 + 4aU}}{2a}, \tag{8}$$

where U is the excitation energy of the compound nucleus, which is related to the separation and kinetic energies of emitted neutron and the excitation energy of the residual nucleus by the expression

$$E^* = U - S_n - E_n.$$

Here S_n and E_n are the separation and kinetic energies of the emitted neutron.

If E^* is bellow the particle threshold, it is supposed that the residual nucleus de-excites predominantly by $E1$ gamma-ray emission with an average energy for $(i+1)$th gamma ray:

$$E_{\gamma(i+1)} = 4\left[\frac{E^* - \sum E_{\gamma i}}{a} - \frac{5}{a^2}\right]^{1/2}. \tag{9}$$

Here it is supposed that $E_{\gamma 0} = 0$.

• The γ cascade continues until the residual energy, i.e. $E = E^* - \Sigma E_{\gamma i}$, is smaller than «the γ-ray cut-off region» (E_u, E_d); then the following γ-ray, the so-called «the deciding gamma», is emitted, feeds that states to which the transition has the lowest multipolity. The γ-ray transition probability from states with spin J_i to those with spin J_f is assumed to be

$$P(J_f) \approx \sum P(J_i)\,\rho(J_f). \tag{10}$$

• If the residual energy remains within «the γ-ray cut-off region», partly a subsequent $E1$ γ-ray is emitted and partly «the deciding gamma» is emitted as described above. In this

Table 2. **The isomeric ratio in $^{197}\mathrm{Au}(\gamma,n)^{196m,g}\mathrm{Au}$ photonuclear reaction depending on bremsstrahlung end-point energies**

End-point energy, MeV	Isomeric ratio		
	Present work		Reference
	Experiment	Calculation	
12			$1\cdot10^{-4}$ [13]
14			$5\cdot10^{-4}$[13]
15	$1.28\cdot10^{-4}$	$1.30\cdot10^{-4}$	
16			$1.2\cdot10^{-3}$ [13]
18	$2.94\cdot10^{-4}$	$3.02\cdot10^{-4}$	$2.3\cdot10^{-3}$ [13]
21.5	$3.78\cdot10^{-4}$	$4.10\cdot10^{-4}$	
22			$6.1\cdot10^{-4}$ [20]
23.5	$4.03\cdot10^{-4}$	$4.15\cdot10^{-4}$	$3.0\cdot10^{-3}$ [13]
25			$6.0\cdot10^{-4}$ [19]
50			$5.0\cdot10^{-4}$ [14]
52			$6.10\cdot10^{-4}$ [13] $5.0\cdot10^{-4}$ [15]

case the competing channel with the transition probability $P = (E - E_d)/(E_u - E_d)$ must be taken into account and the isomeric ratios will be calculated by formula

$$R = P\frac{\sum\limits_{J_f=0}^{\text{COS}} P\left(J_f, E - E_d\right)}{\sum\limits_{J_f=\text{COS}}^{\infty} P\left(J_f, E - E_d\right)} + (1 - P)\frac{\sum\limits_{J_f=0}^{\text{COS}} P\left(J_f, E\right)}{\sum\limits_{J_f=\text{COS}}^{\infty} P\left(J_f, E\right)}. \tag{11}$$

In case of bremsstrahlung-induced photonuclear reactions, the isomeric ratios are calculated as the average value:

$$\overline{R} = \frac{\int\limits_{E_{\text{th}}}^{E_m} \phi\left(E_\gamma\right)\sigma\left(\gamma, n\right) RdE}{\int\limits_{E_{\text{th}}}^{E_m} \phi\left(E_\gamma\right)\sigma\left(\gamma, n\right) dE}, \tag{12}$$

where E_{th}, E_m and $\phi(E_\gamma)$ are the reaction threshold, the bremsstrahlung maximum and the continuous photon spectrum respectively, and $\sigma(\gamma, n)$ is the excitation function or cross section of (γ, n) reaction.

The results of the theoretical model calculations of the isomeric ratios are presented in Table 2.

4. RESULTS AND DISCUSSION

In our experiments, the isomeric ratio has been determined from different combinations of gamma spectra in a series of radioactive measurements with the statistical errors less than 0.1% for the isomeric state and 0.06% for the unstable ground state. The typical spectrum of the reaction product ^{196m,g}Au which is produced from the gold sample irradiated with 18-MeV bremsstrahlung for 30 min, measured for 36.06 min after 224-min cooling time, is shown in Fig. 3.

Here we can see that photopeaks 147.73, 168.3 and 188 keV characterizing the isomeric state ^{196m}Au appear against a very high Compton background of photopeaks 333, 355.72 and 426.0 keV characterizing the unstable ground state ^{196g}Au.

On the other hand, the half-life of the isomeric state is significantly shorter than that of the unstable ground state; and while the isomeric state has disappeared completely, the unstable ground state is still very strong. For this reason, the determination of the isomeric ratio in ^{197}Au$(\gamma, n)^{196m,g}$Au photonuclear reaction is very difficult and always of important significance for the nuclear data. In our experiments by choosing optimal sample masses and times of irradiation, cooling and measurement, we succeeded in the determination of the isomeric ratio for the above-mentioned reaction with very low statistical error. The results of our experiments, which have been determined with average error of 5%, are presented in Table 2 and shown in Fig. 4 together with the excitation function taken from [9] and shown in Fig. 5. In Table 2, the results of other authors are also presented for comparison [14–16]. In this table, we can see that our results are in good agreement with those of other authors except for three end-point energies — namely, 16, 18 and 23.5 MeV [13]. On the other hand, we know that for the case of excitation with bremsstrahlung, the isomeric ratio is the average value in the region from the reaction threshold to the end-point energy. Therefore, it is expected that in the GDR the isomeric ratio increases with the increasing end-point

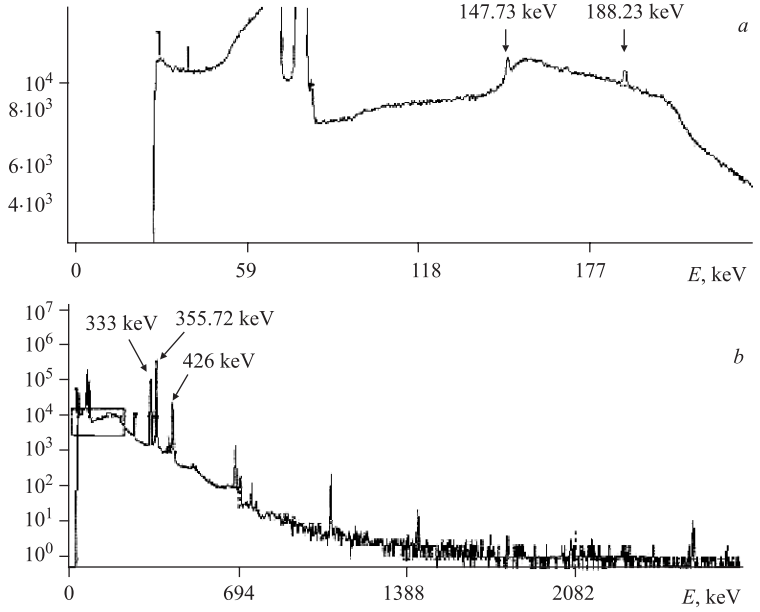

Fig. 3. Spectrum of the gold sample irradiated with 18-MeV bremsstrahlung for 30 min, measured for 36.06 min after 224-min cooling time

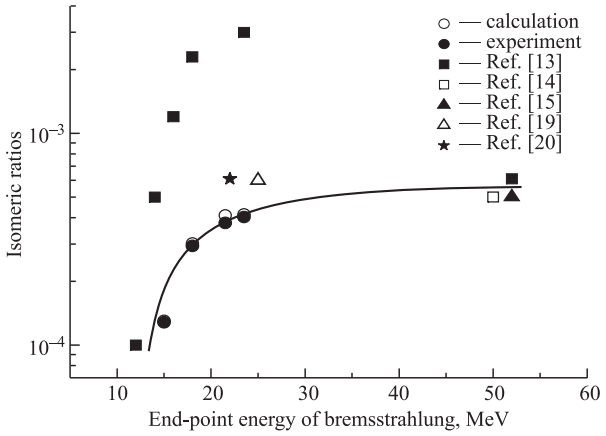

Fig. 4. The dependence of the isomeric ratio on excitation energies in $^{197}\mathrm{Au}(\gamma,n)^{196m,g}\mathrm{Au}$ reactions: ■ the ratio of reaction cross section at a given energy of excitation; ● the ratio of reaction yields, averaged over the whole bremsstrahlung spectrum

energy and reaches maximum value in the end of this region and becomes unchanged for higher energies. As seen from Fig. 5 the isomeric ratio in photonuclear reaction $^{197}\mathrm{Au}(\gamma,n)^{196m,g}\mathrm{Au}$ must have maximum value at about 28 MeV. We also can see from Fig. 4 that for this reaction there is a general trend: in the GDR region, the isomeric ratio increases, reaches maximum value of about 28 MeV and becomes unchanged from the end of this region. This means that our results are completely logical and the isomeric ratios, obtained for 16, 18 and 23.5 MeV by other authors seem to be too high.

We can see that the isomeric ratio is very low. In order to explain this surprising low value, we used the data shown in Table 3 from different references. From this table, we note that in (γ,n) photonuclear reaction, the isomeric ratio depends on the spin of the isomeric and unstable ground states as well as on their difference. For nuclei

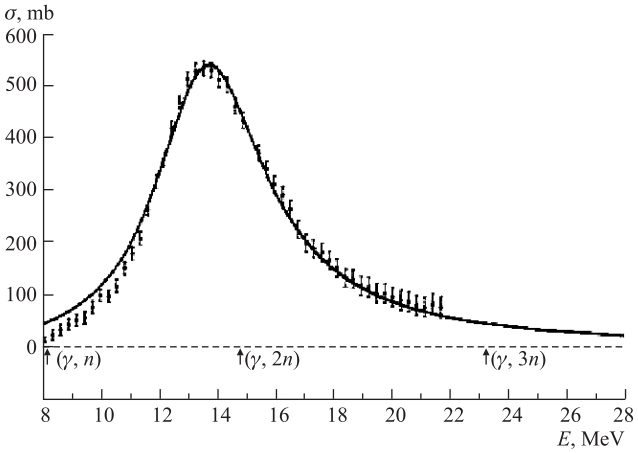

Fig. 5. The excitation function of photonuclear reaction $^{197}\mathrm{Au}(\gamma, n)^{196}\mathrm{Au}$

Table 3. **Isomeric ratios in some (γ, n) photonuclear reactions at 18-MeV bremsstrahlung**

Target nuclei	High spin	Low spin	Spin difference	Isomeric ratio	Reference
^{121}Sb	8^-	1^+	7	0.0516	[5]
^{123}Sb	8^-	2	6	0.0154	[5]
^{86}Sr	$9/2^-$	$1/2^+$	8/2	0.565	[3]
^{142}Nd	$11/2^-$	$3/2^+$	8/2	0.049	[1]
^{144}Sm	$11/2^-$	$3/2^+$	8/2	0.043	[2]
^{90}Zr	$9/2^+$	$1/2^-$	6/2	0.750	[2]
^{110}Pd	$11/2^-$	$5/2^+$	6/2	0.068	[1]
^{140}Ce	$11/2^-$	$3/2^+$	8/2	0.130	[11]
^{138}Ba	$11/2^-$	$3/2^+$	8/2	0.120	[11]
^{136}Xe	$11/2^-$	$3/2^+$	8/2	0.110	[11]
^{197}Au	12^-	2^+	10	$2.94 \cdot 10^{-4}$	Present work
^{130}Ba	$11/2^-$	$1/2^+$	10/2	0.22	[12]
^{132}Ba	$9/2^-$	$1/2^+$	8/2	0.14	[12]
^{134}Ba	$11/2^-$	$3/2^+$	8/2	0.18	[12]
^{136}Ba	$11/2^-$	$3/2^+$	8/2	0.12	[12]

with the same unstable ground state spins, the higher isomeric state spins, the lower isomeric ratios; and for nuclei with the same isomeric state spins, the higher unstable ground state spin, the lower isomeric ratios. In general, the higher differences between the spins, the lower isomeric ratios. For the case of $^{197}\mathrm{Au}(\gamma, n)^{196m,g}\mathrm{Au}$, the isomeric state spin is very high and the difference between the spins is also very high ($\Delta S = 10\hbar$); therefore, the low value of the isomeric ratio is logically proper. Our results are in good agreement with those of other authors except for 16, 18 and 23.5 MeV [13]. On the other hand, from Table 2 we can see that the isomeric ratio changes insignificantly in the giant dipole resonance region. This fact can be explained with the low-momentum transfer in the photonuclear reaction.

In conclusion, we would like to say that the study on the isomeric ratio of the photonuclear reaction $^{197}\mathrm{Au}(\gamma, n)^{196m,g}\mathrm{Au}$ in the GDR region presents an interesting work because of the very high value of the isomeric state spin and the great difference in spins of the isomeric and ground states. The results of the study would contribute to the nuclear data.

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