Partial Photoneutron Cross Sections for the Isomeric State ^{180}Ta^m

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(Received 20 December 2005; published 16 May 2006)

Photoneutron cross sections for ^{181}Ta$(\gamma, n)^{180}$Ta^m were determined from simultaneous measurements of total cross sections ($\sigma^{\rm tot}$) and ground-state cross sections ($\sigma^{\rm gs}$) for ^{180}Ta in photodisintegration of ^{181}Ta with laser Compton-backscattered γ rays. Techniques of direct neutron counting and photoactivation were used for the measurement of $\sigma^{\rm tot}$ and $\sigma^{\rm gs}$, respectively. The partial cross sections for the isomeric state serves as a novel probe of the nuclear level density of ^{180}Ta. Implications for the p- and s-process nucleosynthesis of ^{180}Ta^m are given.

DOI: 10.1103/PhysRevLett.96.192501 PACS numbers: 25.20.−x, 21.10.Ma, 26.50.+x, 27.70.+q

Tantalum-180 is the one and only naturally occurring isomer, as well as the rarest nuclide among all stable nuclei in the solar system. It is present in nature in an isomeric state at the excitation energy of 75 keV with the spin parity of 9^-. The isomer has the half-life greater than 1.2×10^{15} yr, in contrast to the short-lived ground state (1^+: 8.152 h) in ^{180}Ta.

Classically, ^{180}Ta^m is grouped into p-process nuclei [1], where photodisintegration plays a primary role in the presupernova phase of massive stars [2–4] or during their explosions as type-II supernovae [3–7]. Besides the p process, however, it has also been shown that the slow neutron capture process, or s process, in asymptotic giant branch (AGB) stars can be at the origin of a significant production of ^{180}Ta. Cross sections were measured for the ^{181}Ta(γ, n) reaction to constrain the production rate of ^{180}Ta^m [8] in the p process. For the weak branching of the main s-process path to ^{180}Ta^m [9], neutron capture on ^{179}Ta [10] and ^{180}Ta^m [11] and photodeexcitation of ^{180}Ta^m to the ground state through mediating excited states [12] were measured. However, the photodisintegration of ^{180}Ta^m, neutron capture on ^{179}Ta leading to ^{180}Ta^m, and ν_e capture on ^{180}Hf remain experimentally unknown nuclear facets of the p- and s-process origins of ^{180}Ta^m and have to rely on model calculation.

The present study focuses on new photoreaction measurements of the ^{181}Ta$(\gamma, n)^{180}$Ta^m partial cross section that can possibly constrain the nuclear level density (NLD) of ^{180}Ta, a fundamental nuclear parameter in the statistical model which has not undergone a stringent experimental test. In a simple picture of photodisintegration of ^{181}Ta, the $E1$ excitation of ^{181}Ta in the ground state ($7/2^+$) followed by an s-wave neutron emission, ^{180}Ta^m (9^-) is fed in multisteps, for example, by $E1$ transitions, $5^- \to 6^+ \to 7^- \to 8^+ \to 9^-$. Measurements of the partial cross section resulting from this multistep feeding can be a good test to the spin- and parity-dependent NLD of ^{180}Ta at relatively low energy. Partial cross sections were previously provided in a photoactivation measurement with bremsstrahlung [13]. However, this measurement determined the ground-state cross section only and deduced partial cross sections by using the given total cross sections [14]. Photodestruction of ^{180}Ta^m through mediating states [12] may also be a good test to the NLD.

Beams of quasimonochromatic γ rays were produced by means of laser Compton backscattering (LCS) in the electron storage ring Tsukuba Electron Ring for Accelerating and Storage at the National Institute of Advanced Industrial Science and Technology [15]. A diode-pumped Q-switch solid-state laser Nd:YVO_4 provided 532 nm photons at 20 kHz with a doubler module. An external time gate was applied to the laser system to produce LCS γ-ray beams with a repetition scheme of 80 ms beam ON followed by 20 ms beam OFF. The energy spread of the LCS γ-ray beam was 0.8–1.6 MeV in the full width at half maximum.

Two neutron detectors were mounted in the LCS beam line. LCS γ rays passing through a 2 mm diameter lead collimator irradiated tantalum (gold) target materials (ten 1 cm $\times$ 1 cm 100 μm-thick Ta and Au foils, 99.95%) located at the center of the upstream (downstream) neutron detector. Measurements of total photoneutron cross sections ($\sigma^{\rm tot}$) and partial cross sections for the ground state ($\sigma^{\rm gs}$) in ^{180}Ta were simultaneously carried out with techniques of direct neutron counting and photoactivation, respectively. Cross sections for ^{197}Au were measured for a cross-check of the two different techniques.

Photoactivation of the Ta foils was performed at the average γ-ray energies of 9.19, 9.73, 10.47, 10.87, 11.54, and 12.29 MeV for 3.3–6.0 hours. LCS γ rays were monitored every ten minutes with a $8'' \times 12''$ NaI(Tl) detector. The number of incident LCS γ rays was determined from pileup spectra measured with the NaI(Tl) detector [8]. The irradiated ten foils were spread onto an acrylic cap that was then attached to the aluminum end cap of a HPGe detector of planer type. The HPGe detector has an active volume of 114 cm^3 (3800 $mm^2 \times 30$ mm). Hafnium x rays from the Ta sample were measured for 24 hours in one-hour intervals, while 355.68 keV γ rays (the $2^+ \rightarrow 0^+$ transition in ^{196}Pt) from the Au sample were measured for 6 days in six-hour intervals.

During the photoactivation, direct neutron counting was carried out in short runs typically for 30 minutes. The neutron detectors are identical, each consisting of three concentric rings of 4, 8, and 8 ^{3}He proportional counters embedded at 38, 70, and 100 mm, respectively, from the beam axis in a polyethylene moderator. The neutron detection efficiency of the triple-ring detector was calibrated with a ^{252}Cf source. The average neutron energy was determined based on the ring-ratio technique [8,16].

Besides the photoactivation measurement, total cross sections were separately measured at 9 average γ-ray energies over 10.1–12.6 MeV with Ta and Au target samples of 4 mm thickness. Details of the experiment will be described elsewhere.

In the present direct neutron counting, the energy integral of the product of the energy distribution of the LCS γ rays $[n_\gamma(E)]$ and the total photoneutron cross section $[\sigma^{\rm tot}(E)]$ above the neutron separation energy (S_n) was determined from

$$\int_{S_n} n_\gamma(E)\sigma^{\rm tot}(E)dE = \frac{n_n}{n_t f \epsilon_n}. \quad (1)$$

This energy integral is experimentally determined from the number of neutrons n_n, the number of target nuclei per unit area n_t, the correction factor for γ attenuation in thick-target measurements f, and the neutron detection efficiency ϵ_n. Note that $f = (1 - e^{-\mu t})/(\mu t)$, where μ is the linear attenuation coefficient and t is the thickness of the target material.

The total cross section $\sigma^{\rm tot}(E)$ was determined in the energy region above 10 MeV by a least-squares fit to the experimental values of the energy integral assuming the Lorentz line shape.

$$\sigma^{\rm tot}(E) = \sum_{k=1}^{2} \frac{\sigma_k}{1 + [(E^2 - E_k^2)^2/E^2\Gamma_k^2]}, \quad (2)$$

where σ_k is the peak cross section at the resonance energy E_k, and Γ_k is the full width at half maximum. The best-fit Lorentz parameters are listed in Table I.

Figure 1 shows a typical x-ray energy spectrum measured with the HPGe detector along with the time variation of the x-ray yield. $K\alpha_1$ (55.79 keV) and $K\alpha_2$ (54.61 keV) x rays were identified with the half-life corresponding to that of ^{180}Ta$^{\rm gs}$.

In the photoactivation measurement for Ta, the energy integral for the ground-state cross section $(\sigma^{\rm gs})$ was determined from

$$\int_{S_n} n_\gamma(E)\sigma^{\rm gs}(E)dE = \frac{Ye^{\lambda t_{\rm ir}}}{n_t g h \epsilon_X (e^{-\lambda t_i} - e^{-\lambda t_f})} \times \frac{\int_0^{t_{\rm ir}} K(t)dt}{\int_0^{t_{\rm ir}} e^{\lambda t} K(t)dt}. \quad (3)$$

Here Y is the total yield of Hf x rays, λ is the decay constant of ^{180}Ta$^{\rm gs}$, $t_{\rm ir}$ is the irradiation time, $K(t)$ is the time variation of the LCS γ-ray intensity normalized to unity, t_i and t_f are the start and stop time, respectively, for the radioactivity counting, ϵ_X is the detection efficiency for x rays, g is the emission rate of Hf x rays per decay of ^{180}Ta$^{\rm gs}$, and h represents a correction for the self-absorption of x rays in the activated target foils. The K x-ray emission rates were calculated following Ref. [19]; g is 33.1% for $K\alpha_1$ and 19.2% for $K\alpha_2$. The detection efficiency was calibrated with standard sources of ^{109}Cd (88.04 keV) and ^{241}Am (59.54 keV) placed at the same position as the activated foils. The sum effect for the γ rays and associated x rays was negligibly small. The dependence of the efficiency on photon energy was obtained from an EGS4 simulation [20] normalized to the data.

The ratio (η) of the integral for the ground-state cross section [Eq. (3)] to that for the total cross section [Eq. (1)] was determined from the experimental quantities on the right-hand sides of the two equations. From the ratios and the total cross section given in Table I, the ground-state cross sections were determined. Table II lists results of the total, the ground-state, and the isomeric-state cross sections for the Ta sample at the average γ-ray energies. Results of the direct neutron counting and the photoactivation agreed with each other for the Au sample

TABLE I. The best-fit Lorentz parameters for the total cross section. σ parameters of Ref. [17] are multiplied by a factor of 1.22 following the recommendation of Ref. [18].

Experiment	E_1 (MeV)	σ_1 (mb)	Γ_1 (MeV)	E_2 (MeV)	σ_2 (mb)	Γ_2 (MeV)
Present	12.60	264	2.37	15.09	374	4.72
Bergère [14]	12.30	259	2.43	15.23	341	4.48
Bramblett [17]	12.54	188	1.67	14.95	339	5.23

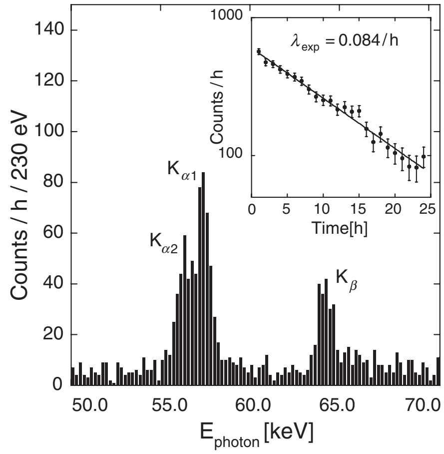

FIG. 1. Typical K x-ray spectrum and the time variation of the $K\alpha$ yield in the photoactivation of the Ta sample.

($\eta = 0.99 \pm 0.015$). Statistical uncertainties are given in the parentheses in Table II.

The systematic uncertainty for the total cross section is 4.4% (3% for n_γ and 3.2% for ϵ_n), while that for the ground-state cross sections is 3% (ϵ_X) if a common source of the uncertainty (n_γ) is eliminated due to the simultaneous irradiation. These two uncertainties are independently propagated to the systematic uncertainty for σ^m if they are uncorrelated. However, the η value obtained for ^{197}Au shows that the neutron and the photon efficiencies are correlated in such a way that their ratio is close to the true value with high precision. This correlation is characteristic of the present detection system essentially independent of photon and neutron energies. The systematic uncertainties for σ^m and σ^{gs} were estimated from error propagation in $\sigma^m = \sigma^{tot}$ (1-η) and $\sigma^{gs} = \eta\ \sigma^{tot}$ with $\Delta\sigma^{tot}/\sigma^{tot} = 4.4\%$ and $\Delta\eta/\eta = 1.5\%$, respectively. The uncertainty for σ^m is 10%–26% as given in the brackets in Table II and is 4.6% for σ^{gs}.

The weak partial cross section ^{181}Ta(γ, n)^{180}Ta^m is sensitive not only to the ^{181}Ta E1-strength function and ^{180}Ta - neutron optical potential (in a similar way as the total photoneutron cross section) but also to the detailed level spectrum in ^{180}Ta responsible for the E1 photon cascade down to the 9^- isomeric state at energies typically below some 5 MeV for the photon energies considered here. To study the partial cross section, use is made of the TALYS nuclear reaction code [21]. Of particular interest here is the prescription used to estimate the NLD. Two microscopic models are considered. The first model uses a statistical calculation that takes into account the discrete structure of the single-particle spectra associated with the Hartree-Fock BCS potential [22]. The second NLD model considered here is based on the combinatorial approach using the single-particle scheme and pairing strength derived from a Hartree-Fock-Bogoliubov calculation [23,24]. These two models correspond to the only microscopic calculation capable of reproducing experimental s-wave neutron spacings with an accuracy comparable to the one obtained with the analytical parametrized formula of the backshifted-Fermi gas type. The latter model has the major advantage over the statistical model of predicting not only the parity dependence but also any nonstatistical limit at low energies.

The photoreaction cross sections predicted with both NLD models are compared with experimental data in Fig. 2. The E1-strength function is adjusted to reproduce the total photoneutron cross section which is insensitive to the adopted NLD model. In contrast, the partial cross section for the isomeric state is relatively sensitive to the NLD as expected, and both NLDs give rise to significant differences which essentially originate from the large NLD obtained with the combinatorial approach for spins $J > 5$ at low energies. In this case, the number of intermediate states capable of feeding the isomeric 9^- state increases and favors the isomeric channel. The combinatorial approach which does not assume a Gaussian spin distribution at low energies (in contrast to the statistical approach) is seen to be in close agreement with the measurement.

A least-squares analysis of the experimental data using the statistical and systematic uncertainties combined linearly showed that the associated error in NLD which corre-

TABLE II. Present results of total photoneutron cross sections and cross sections for the ground state (1^+) and the isomeric state (9^-) in ^{180}Ta in the disintegration of ^{181}Ta.

E_γ^{av} (mb)	σ^{tot} (mb)	σ^{gs} (mb)	σ^m (mb)
9.2	48(0.9)	45(3.1)	3(3.2)[0.7]
9.7	73(0.7)	66(3.8)	6(3.9)[0.9]
10.5	109(1.3)	103(2.6)	6(2.9)[1.6]
10.9	143(1.7)	130(3.7)	13(4.1)[2.0]
11.5	234(1.8)	219(4.9)	15(5.2)[3.4]
12.3	383(27)	329(10)	54(29)[5]

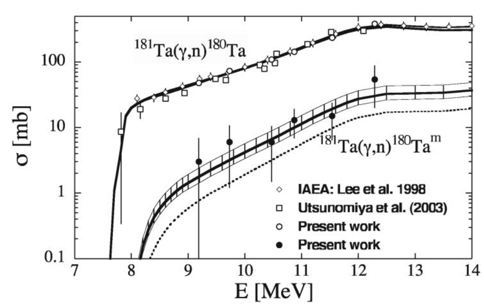

FIG. 2. Comparison between experimental and theoretical cross sections. Open symbols correspond to the total photoreaction [8,18] and solid symbols to the partial cross section for the isomeric state only. The overall (statistical plus systematic) uncertainty is shown on the partial cross section. The solid (dashed) line is obtained with the combinatorial (statistical) NLD.

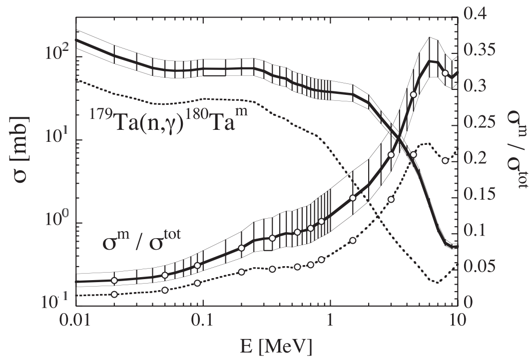

FIG. 3. Predicted ^{179}Ta$(n, \gamma)^{180}$Ta^m cross sections using the combinatorial (solid line) and statistical (dashed line) NLD. The ratio of the partial cross section to the isomeric state to the total cross section is also shown (line with symbols; right axis).

sponds to an increase of 1 from the χ^2 minimum (1.32) is $\pm 28\%$ with the χ^2 probability 93%. We show in Fig. 2 error bars on the theoretical prediction obtained in the least-squares analysis that correspond to $+34\%$ and -22% variations from the default ^{180}Ta NLD for spin $J > 5$ (i.e., $\pm 28\%$ around the mean value).

For nucleosynthesis applications, the new measurements provide some significant constraints on the ^{180}Ta NLD. Such data do not affect the total ^{181}Ta photoneutron cross section of interest in the p-process nucleosynthesis. Indeed, the p process takes place at relatively high temperatures ($1.8 \leq T\,[10^9\text{ K}] \leq 3$), which ensure the thermal equilibration between the ground and isomeric states in the photodestruction of ^{180}Ta as well as in its production by the (γ, n) reaction on ^{181}Ta or the ν_e capture on ^{180}Hf [1,6,7].

In contrast, during the s-process nucleosynthesis, ^{180}Ta^m can be synthesized through the ^{179}Hf$(\beta)^{179}$Ta$(n, \gamma)^{180}$Ta^m branching [9]. The ^{180}Ta^m production depends on the partial neutron cross section for the isomeric state which is found to be sensitive to the adopted NLD prescription. To illustrate such an impact, we show in Fig. 3 the ^{179}Ta$(n, \gamma)^{180}$Ta^m cross sections as well as the isomer-to-total cross section ratio obtained with both NLD prescriptions. The importance to describe properly the low-energy NLD for high spins in this specific reaction channel is illustrated by the error bars around the combinatorial prediction as in Fig. 2. At 30 keV, we find $\sigma^m = 90 \pm 22$ mb and $\sigma^m/\sigma^{\text{tot}} = 0.04 \pm 0.01$. The cross section is twice as large as the theoretical prediction of Ref. [25], whereas the branching ratios are in good agreement. It is to be noted that additional uncertainties still affect our understanding of the s-process origin of ^{180}Ta^m. These concern, in particular, the ^{179}Hf β-decay rates in stellar conditions and the detailed temperature history in which the s process takes place in AGB stars.

In summary, the partial cross section for the isomeric state (9^-) in photodisintegration of ^{181}Ta, which represents selective γ transitions between relatively high-spin states, served as a novel probe of the low-energy NLD of ^{180}Ta at high spins ($J > 5$). Based on the present experiment, the impact of the combinatorial ^{180}Ta NLD on the ^{179}Ta$(n, \gamma)^{180}$Ta^m cross section was analyzed in the context of the s-process origin of ^{180}Ta^m. There would be many cases to study NLD by measuring partial cross sections for isomeric states, e.g., in ^{195}Pt, ^{186}Re, ^{177}Hf, ^{176}Lu, etc.

The authors thank T. Hayakawa for the loan of the γ calibration sources from JAEA. This work was supported by the Japan Private School Promotion Foundation and the Japan Society of the Promotion of Science. S. G. acknowledges the financial support of the FNRS (Belgium).

[1] M. Arnould and S. Goriely, Phys. Rep. **384**, 1 (2003).
[2] M. Arnould, Astron. Astrophys. **46**, 117 (1976).
[3] M. Rayet *et al.*, Astron. Astrophys. **298**, 517 (1995).
[4] T. Rauscher *et al.*, Astrophys. J. **576**, 323 (2002).
[5] S. E. Woosley and W. M. Howard, Astrophys. J. Suppl. Ser. **36**, 285 (1978).
[6] S. E. Woosley *et al.*, Astrophys. J. **356**, 272 (1990).
[7] S. Goriely *et al.*, Astron. Astrophys. **375**, L35 (2001).
[8] H. Utsunomiya *et al.*, Phys. Rev. C **67**, 015807 (2003).
[9] K. Yokoi and K. Takahashi, Nature (London) **305**, 198 (1983).
[10] M. Schumann and F. Käppeler, Phys. Rev. C **60**, 025802 (1999).
[11] K. Wisshak *et al.*, Phys. Rev. C **69**, 055801 (2004).
[12] D. Belic *et al.*, Phys. Rev. Lett. **83**, 5242 (1999).
[13] A. P. Tonchev and J. F. Harmon, Appl. Radiat. Isot. **52**, 873 (2000).
[14] R. Bergère *et al.*, Nucl. Phys. **A121**, 463 (1968).
[15] H. Ohgaki *et al.*, IEEE Trans. Nucl. Sci. **38**, 386 (1991).
[16] B. L. Berman *et al.*, Phys. Rev. **162**, 1098 (1967).
[17] R. L. Bramblett *et al.*, Phys. Rev. **129**, 2723 (1963).
[18] IAEA Report No. IAEA-Tecdoc-1178, 2000.
[19] R. B. Firestone, *Tables of Isotopes* (Wiley, New York, 1996), Appendix F-33.
[20] W. R. Nelson, H. Hirayama, and W. O. Roger, Stanford Linear Accelerator Report No. 265, 1985.
[21] A. J. Koning, S. Hilaire, and M. C. Duijvestijn, in *International Conference on Nuclear Data for Science and Technology*, edited by R. C. Haight *et al.*, AIP Conf. Proc. No. 769 (AIP, New York, 2005), p. 1154.
[22] P. Demetriou and S. Goriely, Nucl. Phys. **A695**, 95 (2001).
[23] S. Hilaire and S. Goriely (to be published).
[24] S. Hilaire *et al.*, Eur. Phys. J. A **12**, 169 (2001).
[25] Zs. Németh *et al.*, Astrophys. J. **392**, 277 (1992).