Formation of isomers in the reaction $^{238}U(p,3n)^{236}Np$

J. Aaltonen
Department of Radiochemistry, University of Helsinki, 00170 Helsinki, Finland

M. Brenner
Department of Physics, Åbo Akademi 20500 Turku, Finland

S. A. Egorov, A. M. Fridkin, V. B. Funschtein, and E. A. Gromova
V. G. Klopin Radium Institute, Leningrad, U.S.S.R.

P. Manngård
Department of Physics, Åbo Akademi 20500 Turku, Finland

V. A. Rubchen'a, Yu. A. Selitsky, and V. A. Yakovlev
V. G. Klopin Radium Institute, Leningrad, U.S.S.R.

V. S. Zenkevich
I. V. Kurchatov Atomic Energy Institute, Moscow, U.S.S.R.
(Received 7 March 1989)

The energy dependence of the production cross sections in long-lived (l) and short-lived (s) isomeric states in ^{236}Np together with the isomeric ratio of the reaction $^{238}U(p,3n)^{236}Np$ near the reaction threshold was measured. The yield of $^{236}Np^s$ was determined from the alpha activity of ^{236}Pu and the yield of $^{236}Np^l$ from analysis by thermal neutron fission. From the energy dependence of the isomeric ratio it follows that the long-lived state is the ground state in ^{236}Np. Theoretical calculations of the yields support these results.

INTRODUCTION

It is known that ^{236}Np has two isomeric states: the long-lived $^{236}Np^l$ with the half-life 1.55×10^5 yr and the spin parity 6^-,[1] and the short-lived one, $^{236}Np^s$ with the half-life 22.5 h and the spin parity 1^-.[2] Hitherto it has not been experimentally established which of these two is the ground state of ^{236}Np. It has been assumed that $^{236}Np^s$ is the ground state.[3-5] In directories[6,7] no indication at all is given as to which is the ground state. It follows from the systematics of the energies of the isomeric states that the ground state of ^{236}Np must be the long-lived isomeric state, and that the short-lived isomer has an excitation energy of 50 keV.[8] The upper limit of the excitation energy of $^{236}Np^s$ was calculated to be 150 keV.[1] This evaluation was made using the single-particle model and taking into account the Gallagher-Moshkovskiy rule. The scheme for the low-lying levels of ^{236}Np has been calculated taking into account the residual p-n interaction[9,10]; values of 77 ± 5 and 190 keV, respectively, were obtained for the lowest excitation energy of ^{236}Np. As can be seen from this, at present there is neither unambiguous evidence as to which isomer is the ground state nor has a single value for the excitation energy of the metastable state been determined.

One of the ways to evaluate the excitation energy of the metastable state is to determine the energy dependence of the isomeric ratio near the threshold of the reaction at which ^{236}Np is formed. Thus, when the energy of the incident particle is lowered to the threshold value, the isomeric ratio $R=\sigma^s/\sigma^l$ must tend to zero if the ground state of ^{236}Np is long lived, and to infinity if it is short lived. Furthermore, as will be shown below, the rate at which R changes depends on the value for the excitation energy of the metastable state.

Isomeric ratios of ^{236}Np have been measured for the following reactions: $^{235}U(d,n)$,[11] $^{238}U(d,4n)$,[5,12,13] $^{237}Np(n,2n)$,[14] and $^{238}U(p,3n)$.[15] All of these, with the exception of the last one, however, were carried out at the energies of the initiating particles, which considerably exceeded the threshold for the ^{236}Np formation reaction. When the isomeric ratios of ^{236}Np in the reaction $^{238}U(p,3n)^{236}Np$ were determined at $E_p=13-30$ MeV,[15] the technique used did not permit a detailed determination of the behavior of the isomeric ratio near the threshold.

The aim of the present work was to study the energy dependence of the cross section of the formation of the long-lived (σ^l) and the short-lived (σ^s) states of ^{236}Np and the isomeric ratio of the reaction $^{238}U(p,3n)^{236}Np$ near the threshold ($E_{\text{thresh}}=13$ MeV). In addition, the purpose was to determine the characteristics of the ground state, the excitation energy of the metastable state of ^{236}Np, and the formation mechanism of the isomeric states in a multicascade process. Studying the isomeric ratio of the reaction $^{238}U(p,3n)^{236}Np$ near the threshold makes it possible to study the joint effect of the threshold and the Coulomb barrier ($E_c=15$ MeV) on the energy dependence of R.

EXPERIMENT

Irradiation of uranium targets was carried out with the MGC-20 cyclotron of Åbo Akademi. Six targets of isotopically pure ^{238}U, each with a thickness of approximately 1.5 mg/cm^2 mounted in sandwich on aluminum backings 0.1 mm thick were irradiated with a 10-μA proton beam with $E_p = 17$ MeV for 100 h. The accumulated proton charge was measured with a Faraday cup with an error of 0.5%.

The number of ^{236}Np^s nuclei formed was determined from the alpha activity of the daughter nuclei ^{236}Pu ($T_{1/2} = 2.851 \pm 0.008$ yr).[8] The alpha activity of the irradiated targets was measured with a surface-barrier silicon detector. It was necessary to take into account the fact that not only ^{236}Pu but also alpha active ^{238}Pu from the reaction

$$^{238}\mathrm{U}(p,n)^{238}\mathrm{Np} \xrightarrow{\beta^-} {}^{238}\mathrm{Pu}$$

accumulated in the target.[16] Because of the considerable thickness of the uranium targets (1.5 mg/cm^2), it was impossible to resolve the ^{236}Pu and ^{238}Pu peaks in the alpha spectra. The relative ratio of the activity of the plutonium isotopes was obtained by measuring the alpha spectra of the aluminum foils separating the uranium targets during proton irradiation. Uranium fission fragments partly sputtered the target's material; this caused very thin layers to form on the aluminum foils and resulted in very good resolution of the ^{236}Pu and ^{238}Pu peaks in the alpha spectra of the layers. This method made it possible to measure the reaction cross sections of both ^{238}U$(p,n)^{238}$Np (Fig. 1) and ^{238}U$(p,3n)^{236}$Np^s (Fig. 2).

A high-sensitivity method to determine the number of ^{236}Np^l nuclei in highly enriched ^{238}U targets by means of thermal neutron fission on the basis of the known fission cross section[17] was used. In the first stage of determining the amount of ^{236}Np^l in the proton irradiated uranium targets, use was made of their isotopic purity. The six targets with the ^{236}Np^l to be determined and analogous uranium targets not subjected to preliminary proton irradiation were irradiated simultaneously in the thermal column of the *F*-1 reactor of the I. V. Kurchatov Atomic

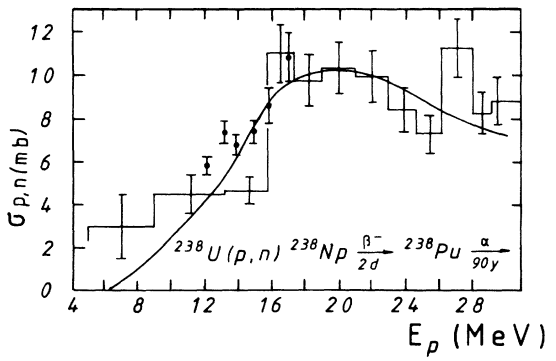

FIG. 1. Cross section of the reaction ^{238}U$(p,n)^{238}$Np. ●, present work. The histogram and curve are from Ageev *et al.* (Ref. 16).

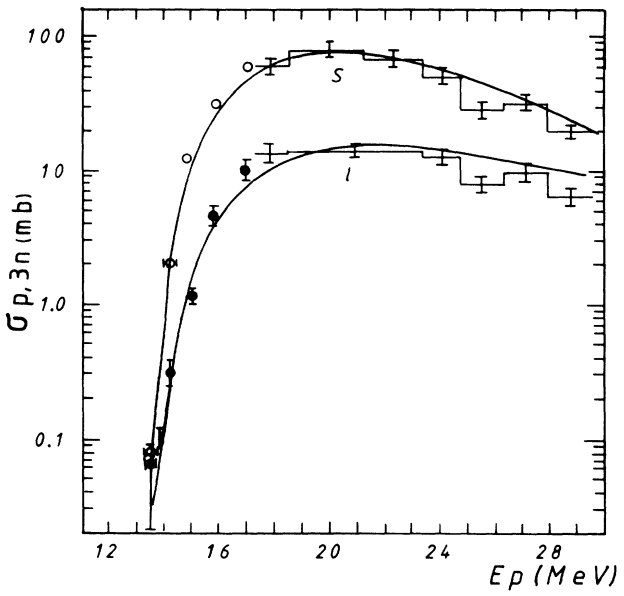

FIG. 2. Cross section of the formation of ^{236}Np isomers in the reaction ^{238}U$(p,3n)^{236}$Np. ○, ^{236}Np^s; ●, ^{236}Np^l. The histograms are from Kovalenko *et al.* (Ref. 15) and Ageev *et al.* (Ref. 16). The curves were calculated.

Energy Institute with a ^{235}U fission cadmium ratio equal to 600. The fission fragments from each of the targets were registered by closely superimposed mica track detectors.[18] The number of ^{236}Np^l fissions was determined as the difference between the total observed number of fissions and the background from the fission of ^{235}U. The number of ^{236}Np^l fissions in the targets ranged from 30% to 6% of the background.

In order to reduce considerably the ^{235}U fission background and to give a more precise determination of the amount of ^{236}Np^l, the neptunium was chemically separated from the uranium targets. Each of the targets was dissolved in concentrated nitric acid. 30–350 ng of ^{237}Np were added to the solutions as a carrier and for determining the chemical yield of neptunium in the chemical separation from uranium.

The neptunium was chemically separated from uranium and plutonium by means of extraction chromatography. Finally, electrolytic deposition of neptunium was made on a stainless-steel backing. The neptunium yield was 80%. To determine the amount of ^{236}Np^l, the targets with mica detectors to detect fission fragments were irradiated with thermal neutrons, as just described. The measured and calculated cross sections of ^{236}Np^l formation and the isomeric ratios are presented in Figs. 2 and 3. A uranium target irradiated with protons at $E_p = 12.5$ MeV (lower than ^{238}U$(p,3n)^{236}$Np reaction threshold) was used for background determination. The background level at $E_p = 13.5$ MeV was equal to 70% of the full number of fission events.

The calculations were made on the basis of the STAPIF code[19] using the Hauser-Feschbach model, as in previous work.[16] To calculate the cross section of ^{236}Np isomeric

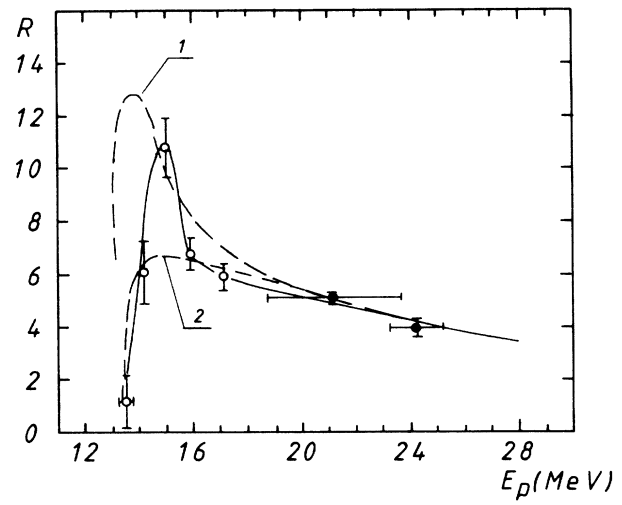

FIG. 3. Isomeric ratio $R=\sigma^s/\sigma^l$ for the $^{238}U(p,3n)$ reaction. ○, present work; ●, Kovalenko *et al.* (Ref. 15). Solid line, experiment. Dashed lines, results of calculations with the spectrum of Fig. 4, $\Delta E_{is}=70$ keV (1), and with "shifted spectrum," $\Delta E_{is}=170$ keV (2).

state formation more exact information is needed about the sequence of the ^{236}Np bandhead energies. The energies of the low-lying levels in the odd-odd ^{236}Np nucleus were calculated from the model of residual interaction between odd particles similar to the way calculated for ^{236}Np by Sood.[10] The main differences between our calculations and those of Sood are that we did not use experimental spectra but one-quasiparticle spectra, which were calculated within the Bardeen-Cooper-Shreiffer (BCS) approach for neutron and proton systems in order to take into account the ground-state formation difference for neighboring nuclei. We also used the finite-range neutron-proton interaction including the Wigner and the spin-spin interaction terms. Part of the calculated two-particle state scheme in ^{236}Np is presented in Fig. 4. Our calculated energy distance between the isomeric states ΔE_{is} is about half that of Sood[10] and the spectrum of two-particle states is more dense. For the calculation a spectrum of 100 states was used.

To calculate the γ-ray cascade after neutron evaporation we have assumed the cascades to occur within the rotational bands.[9] Then transitions to the other bandheads with $K=0,\pm1$ were taken into account with branching ratios depending upon the γ-ray energy. Calculations of the cross section σ were made, including the fission channel at each stage of the neutron cascade.

DISCUSSION OF RESULTS

As can be seen from Fig. 3, the isomeric ratio $R=\sigma^s/\sigma^l$ near the threshold increases with increasing E_p and reaches a peak, after which it declines. From this it follows that the ground state of ^{236}Np is the long-lived one. With increasing proton energy the state $^{236}Np^s$ is formed at an increasing rate. A further increase in proton energy above 15 MeV then reduces the R as the introduced angular momentum increases with rising E_p and results in an increased population of $^{236}Np^l$. The dependence of the theoretical results on the excitation spectrum used was examined. The theoretical curve is sensitive not to the sequence of the quasiparticle levels, but to the energy distance ΔE_{is}. In Fig. 3 the results of the isomeric ratio calculations with $\Delta E_{is}=70$ and 170 keV (together with the isomeric states the remainder of the spectrum is shifted) are presented. The difference in R energy dependence is due to the threshold shift and the different rate of the σ^l and σ^s increase in the threshold area (Fig. 2). The performed analysis indicates that the calculated value for $\Delta E_{is}=70$ keV is close to the experimental value.

In conclusion let us compare the isomeric ratio for the $^{238}U(p,3n)$ reaction with known values for R described in the literature and obtained from other reactions where ^{236}Np is formed.[11-14] The results of a comparison of the way in which R depends on the average introduced angular momentum $\bar{I}$ are presented in Fig. 5. The average introduced angular momentum for protons is calculated from the formula $\bar{I}^2=1.9(E_p-10)$, for deuterons from

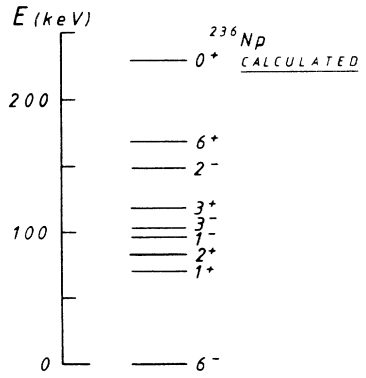

FIG. 4. Calculated bandhead energies for two-particle states in the odd-odd nucleus ^{236}Np.

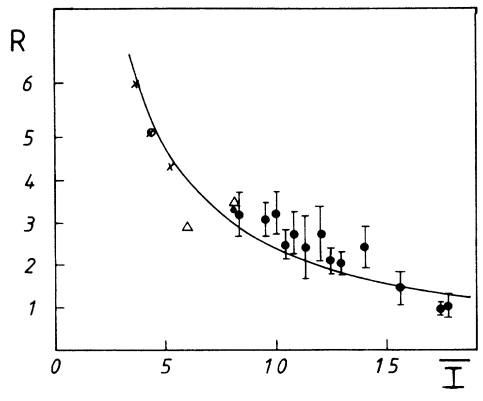

FIG. 5. Dependence of the isomeric ratio R on the average introduced angular momentum $\bar{I}$. ×, $^{238}U(p,3n)$; ○, $^{235}U(d,n)$; ● and △, $^{238}U(d,4n)$; △, $^{237}Np(n,2n)$.

$\overline{I^2}=5.7(E_d-10)$ (Ref. 20) and for neutrons from $\overline{I^2}=2.5E_n$.[21] As can be seen from Fig. 5, for protons and deuterons in reactions with more than one neutron emitted the ratio $R\sim 1/\overline{I}$ (solid curve) proves to be true. The isomeric ratio of the reaction ^{237}Np$(n,2n)^{236}$Np deviates only slightly from the cited relationship for charged particles. More precise consideration of this dependence for different reactions is complicated by the need to take into account the spin of the target nucleus ($I=\frac{5}{2}$ for ^{237}Np), the value for the angular momentum of the evaporated neutrons, and the value for the contribution of direct processes.

[1]M. Lindner, R. J. Dupzyk, R. W. Hoff, and R. J. Nagle, J. Inorg. Nucl. Chem. **43**, 3071 (1981).
[2]C. M. Lederer, J. M. Jaklevic, and S. G. Prussin, Nucl. Phys. **A135**, 36 (1969).
[3]J. H. Landrum, R. J. Nagle, and M. Lindner, Phys. Rev. C **8**, 1938 (1973).
[4]C. K. Paulson and E. J. Hennelly, Nucl. Sci. Eng. **55**, 24 (1974).
[5]P. Lavollee, G. Simonoff, and J. Chenouard, C. R. Acad. Sci. Ser. B **262**, 1739 (1966).
[6]C. M. Lederer and V. Shirley, *Table of Isotopes* (Wiley, New York, 1978).
[7]*International Commission on Radiological Protection,* ICRP Publication 38, Ann. of the ICRP (Pergamon, Oxford, 1985).
[8]M. R. Schmorak, Nucl. Data Sheets **36**, 367 (1982).
[9]D. G. Gardner, M. A. Gardner, and R. W. Hoff, Trans. Am. Nucl. Soc. **47**, 430 (1984).
[10]P. C. Sood, Z. Phys. A **318**, 111 (1984).
[11]J. E. Gindler, J. R. Huizenga, and D. W. Engelkemeir, Phys. Rev. **109**, 1263 (1958).
[12]M. H. Studier, J. E. Gindler, and C. M. Stevens, Phys. Rev. **97**, 88 (1955).
[13]A. Fleury, F. Larrea, and G. Simonoff, C. R. Acad. Sci. Ser. B **271**, 1116 (1970).
[14]W. A. Myers, M. Lindner, and R. S. Newbury, J. Inorg. Nucl. Chem. **37**, 637 (1975).
[15]S. S. Kovalenko, Yu. A. Selitsky, V. B. Funshtein, S. V. Khlebnikov, and V. A. Yakovlev, *Proceedings of the International Conference on Nuclear Data for Science and Technology, Mito,* edited by S. Igarashi (Saikon, Tokyo, 1988).
[16]V. A. Ageev *et al.*, Yad. Fiz. **46**, 700 (1987) [Sov. J. Nucl. Phys. **46**, 700 (1987)].
[17]B. N. Belyaev *et al.*, At. Energ. **60**, 141 (1986).
[18]O. V. Rumyantsev, Yu. A. Selitsky, and V. B. Funshtein, Pribor Technika Experimenta N **1**, 51 (1968).
[19]M. Uhl and B. Strohmaiser, Institut für Radiumforschung und Kernphysik, 76/01, Vienna, 1976.
[20]J. R. Huizenga and R. Vandenbosch, *Nuclear Reactions II,* edited by P. M. Endt and P. B. Smith (North-Holland, Amsterdam, 1962).
[21]I. Halpern and V. M. Strutinski, in *Proceedings of the Second UN International Conference on the Peaceful Use of Atomic Energy 15,* 408 (UN, Geneva, 1958).