Fission isomer of $^{237}Np^m$

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The excitation function and lifetime $T_i = (45 \pm 5)$ ns of a ^{237}Np fission isomer were measured using the $^{238}U(p,2n)$ reaction and the fission-in-flight method. The analysis of the delayed-to-prompt fission ratio provided an excitation energy of (2.85 ± 0.4) MeV for the isomeric state and a branching ratio of 1.9×10^{-3} between γ and fission decays.

NUCLEAR REACTIONS $^{238}U(p, 2n)$; $E_p = 9.75$, 11.6, 12.5 MeV. Measured $^{237}Np^m$ isomeric decay curve and delayed-to-prompt fission ratios. Deduced partial half-lives T_{if}, $T_{i\gamma}$ and the branching ratio.

I. INTRODUCTION

The existence of shape isomers decaying by spontaneous fission,[1-3] the broad maxima in the near threshold cross section for neutron induced fission and (d, pf) reactions,[4] and the evidence of a narrow intermediate structure in the cross sections for subthreshold induced fission,[5] have supplied sufficient proof for Strutinsky's theory of nuclear deformations,[6] which led to the prediction of a second minimum in the potential energy for large values of the deformation.

Subthreshold fission has been interpreted as a coupling of compound nucleus states in the first minimum to intermediate states in the second well, which undergo fission. In the same model a fission isomer represents the ground state in the second well decaying by spontaneous fission through the outer barrier, which may have been populated, for example, after a sequence of neutron evaporations starting from the highly excited compound nucleus formed in the original bombardment. Spontaneously fissioning isomers have been largely studied during the last 15 years and, until now, about 40 fission isomers have been characterized among the heavy elements from berkelium to uranium isotopes.[7]

Isomer half-life and cross section data are normally used to deduce the parameters of the potential barrier to fission.[4, 8-10] In particular, excitation function measurements are needed to deduce from comparisons with models the excitation energy of the fission isomers.[4, 8-9] Systematic investigations of isomeric half-lives have already been carried out and good agreement has generally been reached with theoretical calculations of barrier heights and depths of second minima.[11] However, some discrepancies between theory and experiments do exist; for example, in many isotopes of neptunium the experimental searches for fissioning isomers, contrarily to theoretical predictions, have been unsuccessful.[2, 3, 12, 13]

Several authors suggested that the reason may be found in the γ-ray decay of the Np isomeric states toward the first well. The effect is similar to the one already detected in ^{238}U and ^{236}U,[14, 15] where branching ratios R_i of about 0.1 and 0.2, respectively, were obtained between fission and γ decays of the isomeric states:

$$R_i = \frac{1/T_{if}}{1/T_{if} + 1/T_{i\gamma}} = \frac{T_i}{T_{if}} , \tag{1.1}$$

where T_i, T_{if}, and $T_{i\gamma}$ are the total, partial fission, and partial γ isomeric half-lives, respectively.

Recently Wolf and Unik[16] detected the $^{237}Np^m$, the only known fissioning isomer among neptunium isotopes (half-life 40 ± 12 ns). In their work the delayed-to-prompt fission ratio is reported to be 10^{-8}–10^{-7}, a value quite small when compared with those larger than 10^{-6} observed in other known isomers; the authors analyzed this result with an evaporation model calculation[8] and deduced an isomer excitation energy of 2.7 MeV, assuming barrier parameters for ^{237}Np equal to those of the nearby nuclide ^{238}Np. The evaluated branching ratio ($\sim 7 \times 10^{-4}$) confirms the hypothesis of a prevalent γ decay of the new isomer. In the experiment of Wolf *et al.* the delayed fission activity was electronically measured.

In the present work we aim at confirming the existence of the ^{237}Np isomer, with the use of a different experimental technique; the $^{238}U(p, 2n)$ reaction has been employed as well as a fission-in-flight setup capable to evidence lifetimes and isomeric yields with high efficiencies.[17]

II. EXPERIMENTAL PROCEDURE AND RESULTS

The $^{237}Np^m$ fission isomer was produced by bombardment of ^{238}U with a proton beam from the AVF cyclotron of the University of Milan, extracted with the lowest allowed energy (20 MeV). Since this energy is too high for our purposes, aluminum absorbers of different thicknesses were used to degrade the proton energies in the interval 9.75 to 12.5 MeV. Absorbers were put after the analyzing magnet at the scattering chamber entrance and beams of ~200 nA were obtained. The beam resolution after the analyzing magnet is 20 keV and is brought up to 250 keV by the absorbers as measured by a solid-state detector; some results may be estimated from Ref. 18.

An uranium oxide target, 99,99% ^{238}U enriched, evaporated onto an aluminum backing of 50 $\mu g/cm^2$ was used; the UO_2 oxide layer was 2 mg/cm^2 thick. The range of Np recoils in UO_2 was evaluated by means of Lindhard, Sharff and Schiøtt (LSS)[19] theory: for 60 keV recoils it comes out to be about 110 $\mu g/cm^2$; thus only recoils produced in a region close to the target surface can escape. Experimental escape efficiencies of recoiling Np nuclei in uranium oxide targets have not been measured, but a similar experiment[20] performed with Cm and Am nuclei escaping from Pu and Am targets obtained results in reasonable agreement with LSS theory.

The experimental arrangement is shown in Fig. 1. Nuclei recoiling away from the target undergo fission either in flight or after being stopped in the catcher. Fission fragments emitted backward from these isomers are detected by a thin annular Makrofol foil, 10 μm thick, placed on the target plane. After exposure, the Makrofol foils were

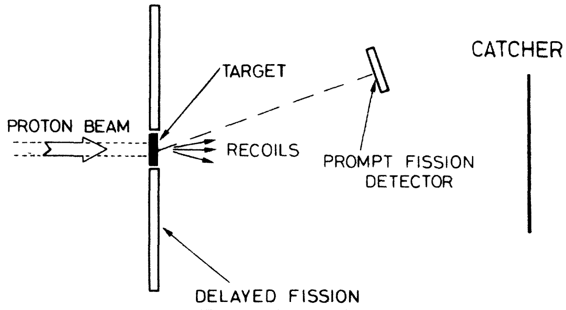

FIG. 1. Sketch of the apparatus for the detection of recoiling fission isomers.

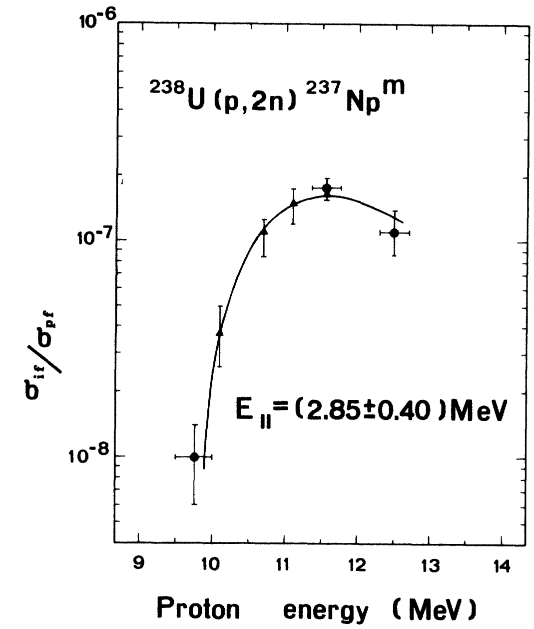

FIG. 2. Delayed-to-prompt fission ratio as a function of the incident proton energies. The curve is from the calculation described in the text. Points represent results of this experiment; triangles are obtained from data in Ref. 2 multiplied by a factor 1.7 and were not used in the fitting procedure.

etched in a 6 N NaOH solution at 60°C for $\frac{1}{2}$ h. The etching develops the fragment tracks into tiny holes optically scanned with a microscope. Prompt fission yields were measured with glass track detectors which allow higher track density in comparison with Makrofol foils. Glasses were placed at several angles with respect to the incident beam direction to take into account the prompt fission fragment angular distributions not completely isotropic at our energies; after exposure, glasses were etched for 1 min in a 40% HF solution and then optically scanned.

The isomeric fission track density distribution versus radial distance from the beam axis, is related[17] to half-lives of the produced isomers; to evaluate the yields, the detection efficiency for our experimental geometry was calculated as in Refs. 17 and 21.

The delayed-to-prompt ratio as measured at three energies shows the typical threshold behavior of a $(p, 2n)$ reaction (full points in Fig. 2); the experimental track density distribution, shown in Fig. 3, indicates therefore the presence of an isomer in the ^{237}Np with a half-life $T_{\frac{1}{2}} = (45 \pm 5)$ ns in good agreement with results in Ref. 16.

Our delayed-to-prompt ratios are a factor 1.7 higher than the ones of Ref. 16; when these data are normalized to ours by means of this factor

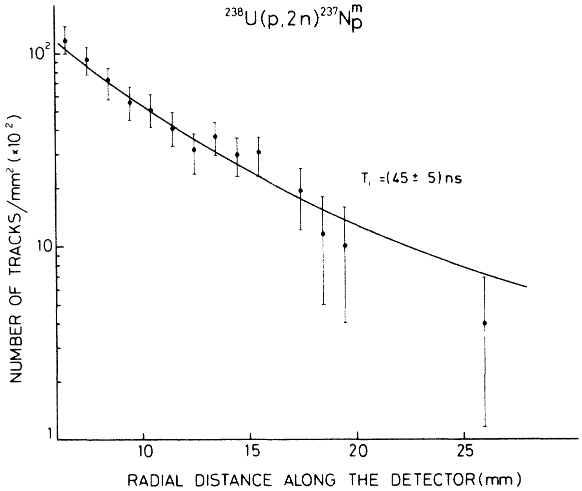

FIG. 3. Decay curve of the $^{237}Np^m$ fission isomer at $E_p = 11.6$ MeV. The points are fitted by a curve obtained with the method described in the Ref. 17.

(full triangles in Fig. 2) the energetic trends of the two measurements are in reasonable agreement.

III. ANALYSIS OF EXPERIMENTAL DATA

The experimental data were analyzed following the statistical model for the $(p, 2n)$ reaction described by Britt *et al.*[4] In the schematic illustration of Fig. 4, A represents the nucleus whose isomer is observed, while $A+1$ and $A+2$ are the nuclei feeding the isomeric state by neutron evaporation. Denoting by Γ_n and Γ_f the neutron and fission decay widths of $A+2$ nuclei, $[\Gamma_n/(\Gamma_n+\Gamma_f)]_{A+2}$ and $[\Gamma_f/(\Gamma_n+\Gamma_f)]_{A+2}$ are the fraction of $A+2$ nuclei forming $A+1$ nuclei and decaying by prompt fission, respectively. The probability that an $A+1$ nucleus is formed at the excitation energy E is given by a Maxwellian distribution:

$$K(E) = \frac{\epsilon}{T^2}\, e^{-\epsilon/T} \tag{3.1}$$

with $\epsilon = E_p + B_p(A+2) - B_n(A+2) - E$, where E_p is the proton incident energy and B_p and B_n are the proton and neutron binding energies of $A+2$ nuclei. T is the nuclear temperature taken as 0.6 MeV. D_I indicates the level spacing of $A+1$ nuclei in the first well at the excitation energy E, Γ_n^I its first class neutron width, and Γ_A^I the probability for the $A+1$ nucleus to go into the second well. For states of the second well D_{II} and Γ_n^{II} have analogous meanings, while Γ_A^{II} gives the probability to go back to first well and Γ_B^{II} the fission probability over barrier B.

All these widths are simply evaluated by the statistical model:

$$\frac{\Gamma_n^I}{D_I} \propto \int_0^{E-B_n(A+1)} \epsilon\rho_I(E - B_n(A+1) - \epsilon)\, d\epsilon \equiv N_I, \tag{3.2}$$

$$\frac{\Gamma_A^I}{D_I} = \frac{1}{2\pi}\int_0^{E-V_A(A+1)} \rho_A(\epsilon)\, d\epsilon = \frac{\Gamma_A^{II}}{D_{II}} \equiv N_A, \tag{3.3}$$

$$\frac{\Gamma_n^{II}}{D_{II}} \propto \int_0^{E-B_n(A+1)-E_{II}(A)} \epsilon\rho_{II}(E - B_n(A+1) - E_{II}(A) - \epsilon)\, d\epsilon \equiv N_{II}, \tag{3.4}$$

$$\frac{\Gamma_B^{II}}{D_{II}} = \frac{1}{2\pi}\int_0^{E-V_B(A+1)} \rho_B(\epsilon)\, d\epsilon \equiv N_B, \tag{3.5}$$

where ρ_I, ρ_{II}, ρ_A, and ρ_B are the density of states at the minima and maxima of the double barrier, V is the barrier height, and E_{II} the isomer excitation energy. In the following calculations the assumption[8]

$$\rho_{II}(\epsilon) = \rho_I(\epsilon - E_{II})$$

has been made while for ρ_A and ρ_B the expressions suggested by Lynn[22] have been used.

Generally in actinide nuclei[23]

$$N_{II} \ll N_A + N_B$$

and, moreover, for $^{238}Np(\equiv A+1)$ $V_B < B_A$ (see for

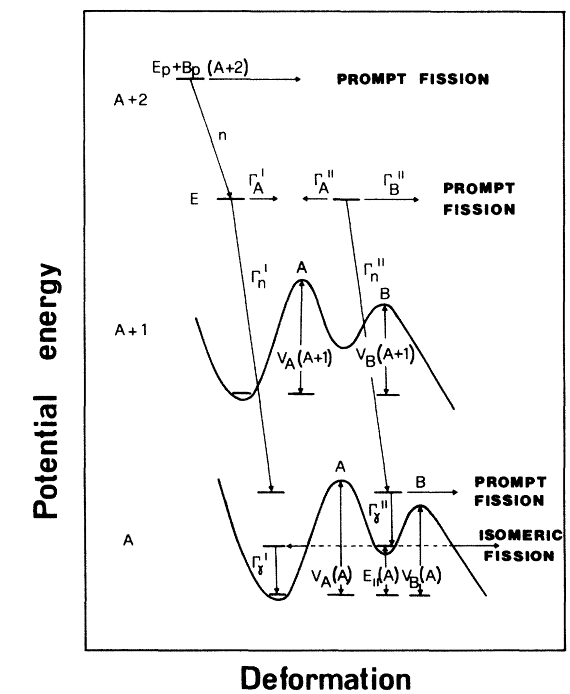

FIG. 4. Schematic illustration of the statistical model used to calculate fission isomer cross section for a $(p, 2n)$ reaction.

TABLE I. Heights and curvatures of the two fission barriers used in the calculation.

Nucleus	V_A (MeV)	$\hbar\omega_A$ (MeV)	V_B (MeV)	$\hbar\omega_B$ (MeV)
^{238}Np	6.19	0.65	5.99	0.45
^{237}Np	5.90	0.80	5.60	0.45

example Ref. 22); therefore it comes out $N_B \gg N_A$.

It follows that the probability a first class $A+1$ nucleus, excited at the energy E, decays to an A nucleus in well I is given by

$$P_{\rm I}(E) \sim \frac{N_{\rm I}}{N_{\rm I}+N_A}\,, \tag{3.6}$$

the probability to decay in well II is given by

$$P_{\rm II}(E) \sim \frac{N_A}{N_B}\,\frac{N_{\rm II}}{N_{\rm I}+N_A}\,, \tag{3.7}$$

and the probability to decay by fission is given by

$$P_f(E) \sim \frac{N_B}{N_{\rm I}+N_A}\,. \tag{3.8}$$

With the assumed approximation the isomeric fission cross section becomes

$$\sigma_{if}(A) = \sigma_c(A+2)\left(\frac{\Gamma_n}{\Gamma_n+\Gamma_f}\right)_{A+2}\left\langle P_{\rm II}(E)\right\rangle_{A+1} \times R_i(A)\,, \tag{3.9}$$

where σ_c is the compound nucleus cross section and the average is performed using the weighting function $K(E)$. The prompt fission cross section is the sum of the following three terms:

$$\sigma_{pf} = \sigma_{pf}(A+2) + \sigma_{pf}(A+1) + \sigma_{pf}(A)\,. \tag{3.10}$$

Since the last contribution is negligible it results in the following:

$$\sigma_{pf} \sim \sigma_c(A+2)\left[\left(\frac{\Gamma_f}{\Gamma_n+\Gamma_f}\right)_{A+2} + \left(\frac{\Gamma_n}{\Gamma_f+\Gamma_n}\right)_{A+2}\left\langle P_f(E)\right\rangle_{A+1}\right].$$

The neutron decay of A into $A-1$, where energetically possible, was taken into account subtracting to the third term in Eq. (3.9) the probability that an A nucleus is formed with an excitation energy exceeding the neutron binding energy. The ratio between Eqs. (3.9) and (3.10) was used to fit our experimental values given in Fig. 2. In the fitting procedure the Γ_n/Γ_f ratio was taken from the systematics of Vandenbosch and Huizenga[24] and the barrier parameters, taken from Ref. 22, are reported in Table I.

The branching ratio R_i in Eq. (3.9) was calculated with Eq. (1.1) imposing to T_i the measured value of 45 ns and to T_{if} the expression

$$T_{if} = \frac{\ln 2}{n\,P_B(E_{\rm II})}\,, \tag{3.11}$$

where $n = \omega_{\rm II}/2\pi$ is the frequency of barrier assaults with $\hbar\omega_{\rm II} = 1$ MeV and

$$P(E) = \left[1 + \exp\left(2\pi\,\frac{V-E}{\hbar\omega}\right)\right]^{-1} \tag{3.12}$$

is the penetrability of an inverted parabolic barrier. The parameter $E_{\rm II}(A)$, as determined by the fit, is equal to (2.85 ± 0.4) MeV, in reasonable agreement with the result (2.7 ± 0.3) MeV of Ref. 16. Inserting this $E_{\rm II}$ value in (3.11) and (1.1) we obtained $R_i = 1.9\times10^{-3}$, $T_{if} = 0.023$ ms and $T_{i\gamma} = 45.1$ ns. The T_{if} value is in good agreement with the 0.01 ms prediction of Ref. 7.

A theoretical evaluation of $T_{i\gamma}$ is more difficult to obtain. A calculation of the γ-branch tried by Lynn[23] in the hypothesis of a small admixture of first with second minimum states, in the limit of complete damping, gives

$$T_{i\gamma} \approx \frac{10^{-5}}{P_A(E_{\rm II})}\,\frac{4D_{\rm I}}{\hbar\omega_{\rm II}}\ {\rm ns}\,. \tag{3.13}$$

Taking the level spacing in the first well to be as in Ref. 22, $D_{\rm I} = 250$ eV, and using the inner barrier parameters from Table I, we obtain $T_{i\gamma} = 250$ ns. The reasonable agreement (approximately a factor 5) with the $T_{i\gamma}$ value deduced from our analysis

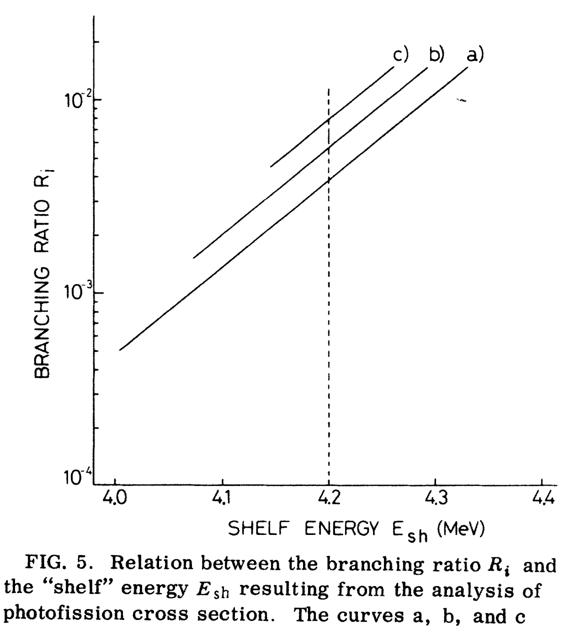

FIG. 5. Relation between the branching ratio R_i and the "shelf" energy $E_{\rm sh}$ resulting from the analysis of photofission cross section. The curves a, b, and c refer, respectively, to $E_{\rm II} = 2.75$, 2.85, and 2.95 MeV.

seems to indicate the consistency of the complete damping hypothesis in the ^{237}Np.

However, the agreement may be fortuitous due to uncertainties in values of V_A, E_{II}, and $\hbar\omega_A$ whose small changes may greatly influence $P_A(E_{II})$.

In a recent work of Bellia *et al.*[25] the branching ratio R_i is deduced from the analyses of deep subthreshold photofission measurements.[26] This study is based on the existence of a "shelf" in the experimental photofission yield[26,27] interpreted in terms of competition between prompt and delayed fission following γ decay in the second well. The photofission cross section may be written as[27]

$$\sigma_{\gamma f}(E)=\sigma_{\gamma f}(\text{prompt})+\sigma_{\gamma f}(\text{delayed})$$

$$=\sigma_{\gamma f}(\text{prompt})\left(1+\frac{R_i P_\gamma(E,E_{II})}{P_B(E)}\right), \quad (3.14)$$

where P_γ, the radiative penetrability in the second well, is given in Ref. 22. Defining the shelf energy E_{sh} as the value where $\sigma_{\gamma f}$(prompt)$=\sigma_{\gamma f}$ (delayed) it results in the following:

$$R_i=\frac{P_B(E_{sh})}{P_\gamma(E_{sh},E_{II})}. \quad (3.15)$$

This relation is shown in Fig. 5, where R_i determined for three values of E_{II} is plotted against E_{sh}. Inserting in (3.15) $E_{sh}=4.2$ MeV (Ref. 25) and $E_{II}=2.85$ MeV, R_i for $^{237}Np^m$ comes out 5.5×10^{-3} in good agreement with our analysis result.

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