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Angular distribution of the photofission fragments of ^{237}Np at threshold energy

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Abstract. Angular distribution of ^{237}Np photofission fragments relative to the photon beam have been measured at energies of 5.60, 6.61 and 8.61 MeV, using an assembly of three independent fission chambers. The 2 MW IEA-R1 reactor at São Paulo was used to obtain the γ rays at these discrete energies following thermal neutron capture in dysprosium, titanium and nickel targets respectively. The experimental results show angular anisotropies of approximately 10% at 5.60 MeV and 6% at 6.61 MeV and an angular distribution practically isotropic (~2%) at 8.61 MeV.

FISSION REACTIONS: ^{237}Np (γ, f), $E_\gamma = 5.60, 6.61, 8.61$ MeV, photofission fragment angular distributions $W(\theta)$.

1. Introduction

It is well known (Bohr 1956) that the spectrum of the excited levels near the fission threshold in a transition state nucleus is expected to be analogous to a normal nucleus near its ground state. Therefore, at low excitation of the transition state nucleus, where a relatively large separation of the levels is expected, only a few of the levels will be available for fission. As the angular distribution of the fission fragments is determined by the quantum numbers of the transition state, it is reasonable to expect to observe anisotropy in the angular distribution through low-lying levels with well defined quantum numbers. Measurements of the angular distribution of fission fragments are thus an important source of information on the nuclear states at the saddle-point configuration. The levels in the transition state nucleus are characterised by the total angular momentum J, its component along the space fixed axis M and the component along the nuclear symmetry axis K.

The angular distribution of photofission fragments for even-A nuclei has been extensively studied (Rabotnov *et al* 1970, Lindgren *et al* 1978, Zhuchko *et al* 1979, Alba *et al* 1981, Ostapenko *et al* 1981, Bellia *et al* 1982). Results of these studies have revealed an energy dependence of the angular distribution in excellent agreement with the hypothesis advanced by Bohr (1956). The ground-state spin of an even-A nucleus is $I_0 = 0$ and the dipole γ-ray absorption produces compound nucleus states $(J, \pm M) = (1, \pm 1)$ with $K = 0, 1$. These K values correspond to the possible open fission channels. At low excitation energies (5–7 MeV) the fission probability is much higher for $K = 0$ channel

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(Bohr 1956); this explains the large angular anisotropies found for even–even nuclei in this energy interval.

On the other hand, photofission of an odd-A nucleus is expected to give an isotropic or nearly isotropic distribution of the fission fragments at energies somewhat higher than the fission barrier. The following factors contribute to this: (i) existence of randomly oriented spin $I_0 \neq 0$; (ii) a large set of available angular momenta J and their components $K \leqslant J$ and (iii) a higher density of open fission channels near threshold. The experimental results (Winhold and Halpern 1956, Baerg *et al* 1959, Katz *et al* 1958) have confirmed this, at least within the quoted errors and at energies greater than 8 MeV. However, as pointed out by Bohr (1956) and Griffin (1959), the best chance of observing the angular anisotropy in an odd-A nucleus would be with ^{239}Pu. The reason for this is that ^{239}Pu has the lowest possible ground-state spin $I_0 = \frac{1}{2}$ and only three degenerate compound nuclear states $(J, \pm M) = (\frac{3}{2}, \pm\frac{3}{2})$, $(\frac{3}{2}, \pm\frac{1}{2})$ and $(\frac{1}{2}, \pm\frac{1}{2})$ are formed by the dipole γ absorption with relative probabilities of $\frac{1}{2}$, $\frac{1}{6}$ and $\frac{1}{3}$, respectively (Griffin 1959). Moreover, it has been pointed out (Bohr 1956, Griffin 1959) that the chances of observing the angular anisotropy should be higher at lower excitation energies. The results of Rabotnov *et al* (1966) and Soldatov *et al* (1970) have in fact shown this to be true using bremsstrahlung γ rays with energies of 5–6 MeV produced by electron accelerators. Values of angular anisotropies as large as 20% were observed in this energy interval for the ^{239}Pu nucleus.

For odd nuclei with spin $I_0 > \frac{1}{2}$, the only indication of an anisotropic angular distribution was reported by Ivanov *et al* (1973) for ^{235}U ($I_0 = \frac{7}{2}$). The measurements were carried out at energies of 6–15 MeV. However, these results were later contradicted by Zhuchko *et al* (1978) who used the same γ source in the same energy range. These authors attributed the previously observed anisotropies in ^{235}U photofission to the presence of impurity of the even–even isotopes of uranium (~10%) in the samples used by Ivanov *et al* (1973). There are no results available for any odd-A nuclei with spin $\frac{5}{2}$ such as ^{237}Np at very low energies. According to Vandenbosch and Huizenga (1973) significant anisotropies at these energies would be expected theoretically (see § 3) if a single particular K state, $K = \frac{5}{2}$ or $\frac{7}{2}$, is the dominant channel in the fission process. In this manner it should be possible to determine the quantum numbers of the lowest lying transition state of these nuclei.

This paper is a continuation of a systematic investigation of photonuclear reactions for ^{237}Np which has been undertaken (Geraldo *et al* 1985) in the energy region near threshold. The main purpose of the present work was to measure the angular distribution of the photofission fragments for ^{237}Np, using monochromatic photons from thermal neutron capture, in the hope of obtaining more accurate distributions at energies lower than those used in the earlier works. The measurements were carried out at three discrete energies, (i) at low energies (5.60 and 6.61 MeV), near the fission threshold of 5.6 ± 0.3 MeV (Vandenbosch and Huizenga 1973, Geraldo *et al* 1985), where the chances of observing some angular anisotropy would be high and (ii) at a reasonably high energy (8.61 MeV), to compare with the results obtained by Baerg *et al* (1959) and Katz *et al* (1958) who used bremsstrahlung photons of 8 MeV.

2. Experimental procedure

Monochromatic γ rays were obtained with an experimental apparatus mounted inside a radial beam hole of the IEA-R1, 2 MW swimming pool type reactor. The γ radiation was produced by the neutron capture in targets when placed near the reactor core, in front of the beam hole. The collimation assembly as well as the special neutron filters were similar to those used by Young and Donahue (1963) and are described in detail elsewhere (Geraldo 1983).

The γ rays used in the present work were obtained by using the targets of Dy, Ti and Ni. The principal γ rays and the most important secondary γ rays, whose line widths are of the order of only a few eV, are listed in table 1 together with their relative intensities (Groshev *et al* 1959). The contributions of these secondary γ rays were taken into account by using the weighted mean of all the γ rays, in a procedure similar to that used by Carvalho *et al* (1963) and Manfredini *et al* (1969). In this way, γ rays with mean energies of 5.60, 6.61 and 8.61 MeV were obtained from the targets of Dy, Ti and Ni respectively.

The design of the experimental apparatus for the angular distribution measurement was constrained by the dimensions, and the amount, of ^{237}Np available. The sample supplied by the International Atomic Energy Agency (IAEA) contained a total of 36.6 mg of ^{237}Np deposited on six titanium discs each with an active diameter of approximately 40 mm, corresponding to a deposit thickness of $\sim$0.5 mg cm^{-2}. An isotopic analysis of this sample by α spectrometry showed only a ^{238}Pu contamination of about 2×10^{-4}%.

The fission chamber assembly shown in figure 1 consists of three independent cylindrical aluminium chambers. It was aligned with the beam hole and positioned at the opposite side to the target element, outside the reactor concrete shield. In this assembly each chamber contained two sample discs joined back to back and suspended by a metallic rod. The discs in three chambers were rotated at approximately 60 RPM during irradiation by an external rotating device in order to avoid the corrections arising from the flat geometry of the samples. The chambers were evacuated to 10^{-3} Torr to ensure that the fission fragments reached the detector. Inside each of these chambers, another cylindrical aluminium tube of 30 cm diameter was placed each with 16 holes of 2 cm diameter at different angles to the sample. These dimensions gave the best combination of fragment intensity at the detector position and smallest solid angle. The fission-fragment detectors were attached to the outside wall of this inner tube, covering the area of each hole. The Makrofol KG (8μ) foils were used to record the fission tracks.

Three fission chambers as opposed to a single one were used in order to obtain the lowest total statistical error in the measurement. In addition, this allowed three independent and simultaneous sets of data which gave a better handle on the systematic uncertainties.

Table 1. Energies and relative intensities of the most important γ rays from the targets used, corrected for any attenuation in the filters of the collimation assembly.

Target	γ rays (MeV)	Relative intensity
Dysprosium	5.58	1.000
	5.89	0.047
Titanium	6.75	1.000
	6.42	0.659
	6.56	0.138
Nickel	8.99	1.000
	8.51	0.397
	8.10	0.086
	7.82	0.200
	7.57	0.124

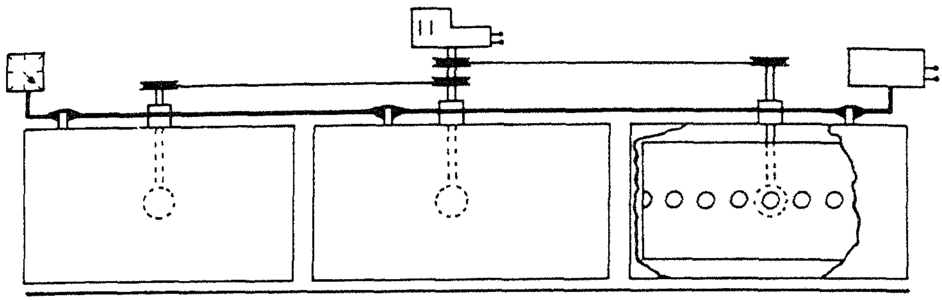

Figure 1. Assembly of the fission chambers used for the angular distribution measurements.

3. Results and discussion

The Makrofol foils were etched in a KOH solution (1.223 g ml^{-1}), at a temperature of 60 °C, for 20 min to make the tracks created by the fission fragments clear. The total number of tracks were counted as a function of angle with respect to the incident photon beam, using an automatic discharge chamber (Geraldo *et al* 1979). Due to the lower fission yield at 5.60 MeV the fission tracks, in this case, were counted visually using an optical microscope (magnification 135). The experimental procedure together with specific details of this technique are described in another paper (Geraldo *et al* 1979).

The background from the (n, f) reaction was measured separately, making irradiations with and without a cadmium foil in the γ beam produced by the Bi target. The capture γ-ray energy of the bismuth target (4.17 MeV) is somewhat below the fission threshold. The (n, f) contribution was found to be negligible.

The number of tracks at corresponding angles were found to be nearly the same in the three chambers, within the experimental errors, and they were added together before the final analysis. The results of the angular distribution are presented in figure 2. The experimental points, shown with error bars, were normalised at 0°. The errors quoted are standard deviations based on the counting statistics. The full curve is the weighted least-squares fit of the experimental data to the function $W(\theta)=a+b\sin^2\theta$. It is clear from the figure that the assumed form of the distribution is consistent with the data in the three cases. The coefficients a and b, normalised such that $\int_0^\pi W(\theta)\sin\theta\,d\theta=1$, are given in table 2 and the angular anisotropies (defined as b/a) are compared with the result of Baerg *et al* (1959) and Katz *et al* (1958) in figure 3. The anisotropies show the expected decrease with increasing energy as in the case of the even-A nuclei. The almost isotropic angular distribution observed at 8.61 MeV is in good agreement with the results of Baerg *et al* (1959) and Katz *et al* (1958) who measured it at 8 MeV.

The theoretical angular distribution for dipole photofission can be represented by the following expression (Soldatov *et al* 1970, Vandenbosch and Huizenga 1973):

$$W(\theta)=\sum_{J,M,K} P(J,M)\frac{2T_f^{J,K}}{\Sigma_{K\leqslant J}\,2T_f^{J,K}+T_c^J}\,W_{M,K}^J(\theta) \qquad (1)$$

where $P(J,M)$ is the relative probability of forming the compound nuclear state (J,M), $T_f^{J,K}$ is the fission transmission coefficient, T_c^J is the combined transmission coefficient $(T_\gamma+T_n)$ of the processes competing with fission and $W_{M,K}^J(\theta)$ is the angular distribution function for the state (J,M,K). The factor of two in the expression accounts for the double degeneracy $(\pm K)$ of the half-integer K values.

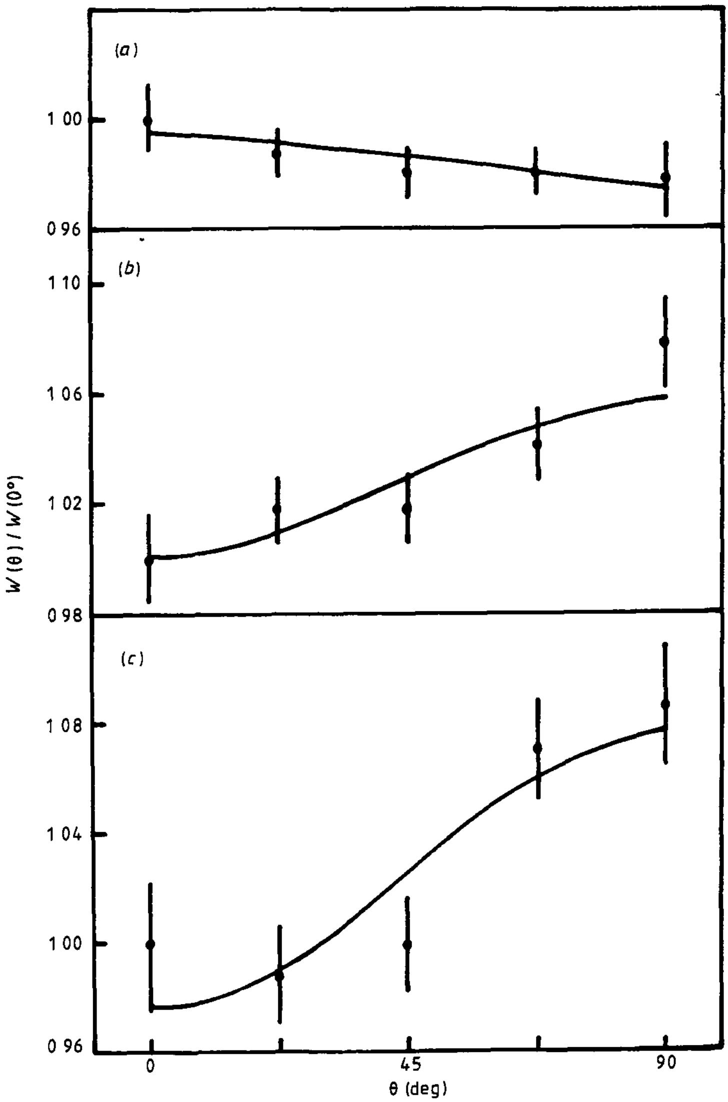

Figure 2. Observed photofission fragment angular distributions for ^{237}Np at energies (*a*) 8.61 MeV, (*b*) 6.61 MeV and (*c*) 5.60 MeV.

Table 2. Values of the coefficients *a* and *b* obtained by fitting the experimental data to the function $W(\theta) = a + b \sin^2 \theta$.

Coefficients	5.60 MeV	6.61 MeV	8.61 MeV
a	0.468 ± 0.008	0.481 ● 0.005	0.506 ● 0.002
b	0.047 ± 0.013	0.027 ± 0.008	−0.009 ± 0.003
b/*a*	0.101 ± 0.027	0.057 ± 0.017	−0.018 ± 0.006

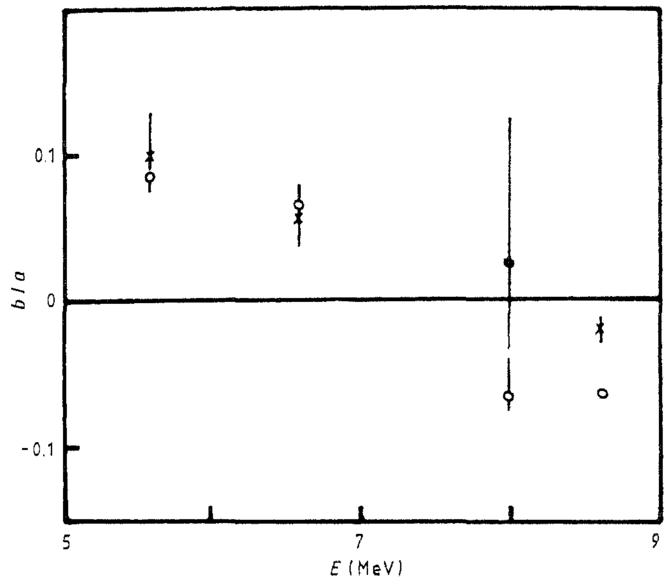

Figure 3. Energy dependence of b/a for photofission of ^{237}Np. ×—results of this work, ●—measurement of Baerg *et al* (1959), O—calculated values of b/a.

The probabilities $P(J, M)$ for different combinations of J and M and the expressions for $W^J_{M,K}(\theta)$ may be found in Katz *et al* (1958), Griffin (1959) and Vandenbosch and Huizenga (1973). Substituting these quantities into equation (1) and keeping in mind that for a target with spin $\frac{5}{2}^+$, $J=\frac{3}{2}, \frac{5}{2}, \frac{7}{2}$; $\frac{1}{2} \leqslant M \leqslant J$ and $\frac{1}{2} \leqslant K \leqslant J$, the following expression is obtained:

$$W(\theta)=\sum_{J,K} (a^{J,K}+b^{J,K}\sin^2\theta)\frac{2T_f^{J,K}}{\Sigma_{K'\leqslant J}2T_f^{J,K'}+T_c^J} \qquad (2)$$

where the values of the coefficients $a^{J,K}$ and $b^{J,K}$ are listed in table 3. The fact that there are four transition states for $J=\frac{7}{2}$, three transition states for $J=\frac{5}{2}$ and two transition states for $J=\frac{3}{2}$ was taken into account by using appropriate weighting factors in this expression.

In a preliminary analysis using formula (2), for the expected angular distribution in the dipole photofission through a specific channel K, some approximations are useful. At energies below the photoneutron threshold the neutron transmission coefficient $T_n=0$. Assuming that $T_f \gg T_\gamma$ and also that the T_f values are independent of J (i.e. the T_f of the rotational members of a particular band are equal) and further, considering that a single transition state K is contributing to fission, the following angular distributions are obtained for $K=\frac{1}{2}, \frac{3}{2}, \frac{5}{2}$ and $\frac{7}{2}$ levels respectively:

$$W_{K=1/2}(\theta)=0.486+0.021\sin^2\theta \qquad b/a=0.044 \qquad (3)$$

$$W_{K=3/2}(\theta)=0.483+0.026\sin^2\theta \qquad b/a=0.054 \qquad (4)$$

$$W_{K=5/2}(\theta)=0.398+0.153\sin^2\theta \qquad b/a=0.385 \qquad (5)$$

$$W_{K=7/2}(\theta)=0.750-0.375\sin^2\theta \qquad b/a=-0.500 \qquad (6)$$

In the approximation of equations (3)–(6) one can see that, theoretically, significant anisotropy may result if a particular K state, such as $K=\frac{5}{2}$ or $\frac{7}{2}$, is the dominant channel in the photofission process near threshold. On the other hand, due to closely similar and very

Table 3. Values of the coefficients $a^{J,K}$ and $b^{J,K}$ calculated for a nucleus with spin $\frac{5}{2}$.

(J, K)	$a^{J,K}$	$b^{J,K}$
$(\frac{3}{2}, \frac{1}{2})$	0.1	0.0166
$(\frac{5}{2}, \frac{1}{2})$	0.2428	−0.1143
$(\frac{7}{2}, \frac{1}{2})$	0.1428	0.1190
$(\frac{3}{2}, \frac{3}{2})$	0.1221	−0.0166
$(\frac{5}{2}, \frac{3}{2})$	0.1857	−0.0286
$(\frac{7}{2}, \frac{3}{2})$	0.1746	0.0714
$(\frac{5}{2}, \frac{5}{2})$	0.0918	0.1837
$(\frac{7}{2}, \frac{5}{2})$	0.3061	−0.0306
$(\frac{7}{2}, \frac{7}{2})$	0.75	−0.375

low theoretical anisotropy expected for $K=\frac{1}{2}$ and $\frac{3}{2}$, it is very difficult to say anything about the dominance of these transition states separately. Thus, in this approximation, the results obtained in this work and presented in table 2 do not allow us to extract information about the individual contributions of the $K=\frac{1}{2}$, $\frac{3}{2}$, $\frac{5}{2}$ or $\frac{7}{2}$ levels. This probably must also occur for other odd-A nuclei with spin $\frac{5}{2}$ or $\frac{7}{2}$ such as ^{233}U, ^{241}Am, ^{243}Am or ^{235}U. However, it might be worthwhile trying to study these nuclei at least at one very low energy (for example 5.6 MeV) in order to verify that the small anisotropies are also obtained in these nuclei.

Even for more realistic calculations with expression (2) it is still useful to make some simplifying assumptions. Assuming a single fission barrier with the shape of an inverted parabola, the fission transmission coefficient is given by (Hill and Wheeler 1953)

$$T_f^{J,K}(E)=(1+\exp[2\pi(E_f^{J,K}-E)/\hbar\omega^{J,K}])^{-1} \qquad (7)$$

where $E_f^{J,K}$ and $\hbar\omega^{J,K}$ are respectively the height and curvature of the fission barrier for the state (J, K) and E is the compound nucleus excitation energy.

The fission barrier heights for the rotational states of each $K=\frac{1}{2}$, $K=\frac{3}{2}$, $K=\frac{5}{2}$ and $K=\frac{7}{2}$ band were calculated with the expression (Bjørnholm and Lynn 1980)

$$E_f^{J,K}=E_f^K+\frac{\hbar^2}{2\mathscr{I}}[J(J+1)-\alpha(-1)^{J+1/2}(J+1/2)\delta_{K,1/2}] \qquad (8)$$

where E_f^K is a constant for a particular K rotational band and represents the variables in expression (2) together with the curvature $\hbar\omega^{J,K}$, $\mathscr{I}$ is the effective moment of inertia and α is the decoupling constant for the $K=\frac{1}{2}$ band. For the calculations, the values of $\hbar^2/2\mathscr{I}$ and α were taken from Bjørnholm and Lynn (1980). According to these authors, in experiments with neutron-induced fission for nuclei in the actinides region, the best values found for these parameters were $\hbar^2/2\mathscr{I}\sim 2.5$ keV and $\alpha\sim 2$.

Whenever necessary the neutron transmission coefficient $T_n(E)$ from the Perey–Buck optical model (Auerbach and Perey 1962) was used. The γ-ray transmission coefficient $T_\gamma(E)$ was taken from Vandenbosch and Huizenga (1973); it is based on a semi-empirical expression for the dependence of the transmission on energy, spin, and the Fermi gas level parameter 'a', normalised to experimental values obtained from resonance capture on several even-A nuclei. Between 4.5 and 6.5 MeV the transmission was approximated by the following empirical equation based on the study of Vandenbosch and Huizenga

$$T_\gamma^{1/2}(E_\gamma)=0.1\exp[(E_\gamma-6.02)/0.41] \qquad (9)$$

where E_γ is in MeV.

Above 6.5 MeV the $T_\gamma^{1/2}$ is a weak function of energy and was considered constant. The T_γ values for other J values were obtained from the relations

$$T_\gamma^{3/2}(E_\gamma)=1.92T_\gamma^{1/2}(E_\gamma) \tag{10}$$

$$T_\gamma^{5/2}(E_\gamma)=2.68T_\gamma^{1/2}(E_\gamma) \tag{11}$$

$$T_\gamma^{7/2}(E_\gamma)=3.25T_\gamma^{1/2}(E_\gamma). \tag{12}$$

In this manner, by using the foregoing equations and a computer search program one may in principle be able to reproduce the experimental values of b/a for ^{237}Np, by adjusting the heights and curvatures of the transition state barriers appropriately. Nevertheless this procedure for calculating the theoretical b/a values is a difficult task because even with the simplifying assumptions mentioned above there are 13 free parameters to be adjusted. By using a further simplifying assumption that $\hbar\omega^{J,K}=\hbar\omega^K$ there are still eight variables and a search can involve a large number of calculations if all the parameters are considered free.

In the present calculations it was further assumed that $\hbar\omega^{J,K}=\hbar\omega$ (Soldatov *et al* 1970, Ostapenko *et al* 1981), thereby reducing the number of free parameters to only five. This assumption, made for the sake of simplifying the calculation, means that there may be many other hypotheses which will fit the experimental data. These five parameters were allowed to vary, within a realistic range ($\hbar\omega=0.1$–1 MeV and $E_f=4$–7 MeV) by using a computer program. Several different sets of these parameters result in equally good fits to the experimental b/a values. However, in all resulting sets $\hbar\omega=0.85$ MeV and $E_f^{7/2}=6.75$ MeV were obtained, showing that these two parameters are rather insensitive to variations of the remaining parameters.

Considering the results $\hbar\omega=0.8$ MeV and $E_f=5.7$ MeV obtained previously (Back *et al* 1973, Geraldo *et al* 1985), the particular set of parameters listed in table 4 was chosen as an illustrative example of such a calculation. Figure 3 shows the theoretical b/a values calculated for $\hbar\omega=0.85$ MeV and the parameters listed in table 4 along with the experimental values. As can be seen, it was possible to reproduce the energy dependence of b/a for ^{237}Np for all energies. The discrepancy between the theoretical and experimental b/a values at 8.61 MeV may be reduced by attributing a different weight factor for the $K=\frac{1}{2}$, $\frac{3}{2}$, $\frac{5}{2}$ and $\frac{7}{2}$ levels at this energy, as has been shown in a similar study for ^{239}Pu (Soldatov *et al* 1970, Vandenbosch and Huizenga 1973). However, in the present case, this procedure was not followed because at this energy the theoretical angular distribution may be considered independent of the fission barrier parameters.

Table 4. Parameters of the transition state barriers obtained in the theoretical calculation of b/a.

K	E_f^K (MeV)
$\frac{1}{2}$	5.25
$\frac{3}{2}$	5.50
$\frac{5}{2}$	5.75
$\frac{7}{2}$	6.75

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