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Observation of new states in the third well of the ^{231}Th fission barrier

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Résumé. — Une mesure simultanée de la probabilité de fission dans la réaction ^{230}Th(d, pf) et de la distribution angulaire des fragments associée révèle la présence de nouveaux états résonnants du thorium-231 aux environs de 5,9 MeV d'énergie d'excitation. Ces états sont interprétés comme étant des membres de haut spin de la paire de bandes de rotation mise en évidence en réaction (n, f). Cette mesure apporte la confirmation de l'existence d'un troisième minimum de la barrière de fission aux déformations octupolaires du noyau ^{231}Th.

Abstract. — A simultaneous measurement of the ^{230}Th(d, pf) fission probability and the associated fission fragment angular distribution reveals the presence of new resonant states of the fissioning ^{231}Th nucleus at excitation energies around 5.9 MeV. These states are interpretated as high spin members of a pair of rotational bands, thus confirming the previous evidence of a third minimum in the fission barrier of ^{231}Th at octupole deformations.

Theoretical and experimental investigations of the so-called thorium anomaly suggested that the potential energy surfaces, associated with the thorium fissioning nuclei, were far more complex [1, 2] than originally assumed. A typical evidence of this anomaly is provided by the ^{231}Th nucleus, where the ^{230}Th(n, f) fission cross section presents a well-known isolated sub-threshold resonance at an incident neutron energy of about $E_n \simeq 720$ keV which corresponds to an excitation energy of $E^* \simeq 5.85$ MeV for the ^{231}Th* nucleus.

This resonance was first interpreted as the manifestation of a rotational band built on a vibrational state located in the second well of a double-humped fission barrier (DHFB) [3]. Such an interpretation requires that the maxima of both the first and the second humps must be higher in energy than the resonant states considered. But for ^{231}Th, the top of the first hump in the fission barrier calculated with the shell correction method [4] lies well below the energy of the resonance in question. Moreover, it also turns out that the measured moment of inertia, $\mathfrak{J}$, is much larger

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than that predicted by the DHFB hypothesis at the second well deformation [5]. Hence, the DHFB interpretation has to be abandoned.

The explanation of this anomaly proposed in terms of a triple-humped fission barrier (THFB) is more convincing. The introduction of mass asymmetric deformations in the evaluation of potential energy surfaces by Möller and Nix [1] leads to second-order modifications of the barrier shapes when the latter are calculated according to the shell correction method. For light actinides, this results in the creation of a third, rather shallow, minimum in the barrier. This minimum is, in fact, double because the pear-shaped (octupole) deformed fissioning nucleus can take two different orientations. This requires the presence of both positive and negative parity rotational bands shifted by an energy value called « inversion energy » [6].

The fine structure components in the ^{230}Th(n, f) fission cross section, observed by Blons *et al.* [7] in the $E_n = 720$ keV region with a resolution of $\Delta E_n = 1.7$ keV, together with experimental results on fission fragment angular distributions (FFAD) and anisotropies [8, 9], did indeed confirm the predicted existence of such a pair of rotational bands with opposite parities.

However, the use of (n, f) reactions limits one to rather small angular momentum transfers of roughly $l \lesssim 3$, so that only spin states $J = 1/2, 3/2$ and $5/2$ are appreciably excited. Moreover, the fine structure components have a width, Γ, of about 7 keV and a mean energy spacing in the $2\text{ keV} \lesssim \Delta E \lesssim 10$ keV range, thus resulting in a certain amount of overlap which can make their ultimate interpretation rather difficult. These drawbacks tend to decrease when using the (d, pf) reaction because, first, one can hope to attain $l \simeq 6$, thus populating spin states of up to $J = 13/2$, and second, the overlap problem ought to be less severe since the rotational level spacing increases with J. Some preliminary evaluations concerning the ^{230}Th(d, pf) reaction thus indicate that the $J^\pi = 9/2^+, 11/2^-$ and $13/2^+$ rotational states ought to be both separately discernible and sufficiently populated to be experimentally accessible.

In this experiment we therefore propose to observe the fine structure in the ^{230}Th(d, pf) cross section at excitation energies, E^*, around 5.9 MeV, together with its associated fission fragment angular distribution. The results will be analysed by means of a simultaneous best fit procedure (SBF), explained in detail in reference [10] where it was applied to similar data obtained from ^{230,232}Th(n, f) reactions [7, 10, 11].

The basic experimental lay-out [12] is schematically represented in figure 1. The $E_d = 12$ MeV deuteron beam was produced by the Saclay tandem Van de Graaff accelerator. The proton energies, E_p, were measured at $\theta_p = 130°$ on the focal surface of a Q3D magnetic spectrometer

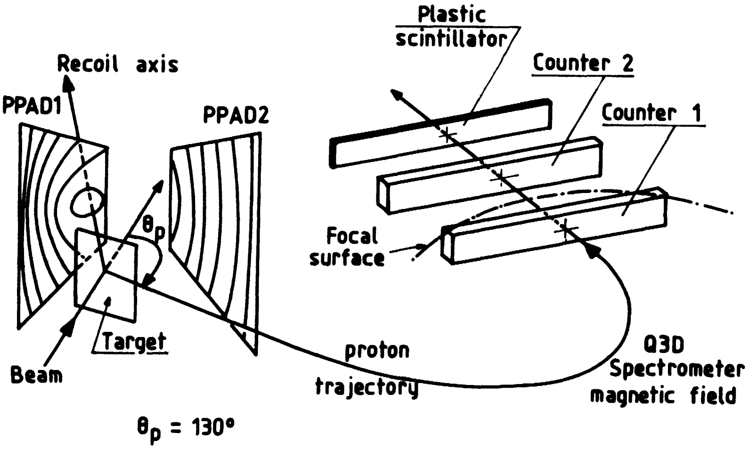


Fig. 1. — Schematic representation of the experimental procedure. PPAD1 and PPAD2 detect the fission fragments and measure the θ angle with respect to the recoil direction. The adjacent strips covering sequential θ widths are separated by an insulator.

by a set of two position-sensitive single-wire proportional counters. The overall energy resolution was $\Delta E \simeq 7$ keV.

Under the above defined experimental conditions, the fission fragment angular distribution exhibits an axial (azimuthal) symmetry with respect to the ^{231}Th*-recoil direction in the laboratory, the latter direction being itself practically independent of E^*, for all E^* of interest in our case. Moreover, since the kinetic energy of the ^{231}Th* recoil is much smaller than the kinetic energy of the fission fragments, the latter will be emitted nearly back to back in the laboratory, thus making the detection of a single fragment at a given angle, θ, sufficient to determine both the fission probability and the fission fragment angular distribution. As shown in figure 1, the two parallel plate avalanche detectors (PPAD), used as fission fragment detectors, are subdivided into adjacent strips covering sequential θ widths so as to cover the $0° \leqslant \theta \leqslant 90°$ range simultaneously.

A (d, pf) event was defined as a fast coincidence between a fission fragment detected by a PPAD and a proton detected by a plastic scintillator set behind the proportional counters. The coincidence window was subsequently reduced by correcting for the proton time of flight, through an identification of the proton trajectory in the Q3D.

One can define an experimental fission probability as being the ratio of the number of detected proton-fragment coincidences (corrected for the PPAD solid angle) to the number of proton counts (corrected for a (d, pn) contribution). A typical set of such fission probability data is shown in figures 2a and 2b for the $0° \leqslant \theta \leqslant 90°$ and $0° \leqslant \theta \leqslant 30°$ intervals respectively. From a qualitative point of view, one can see in figure 2b three resonances at 5.89 MeV, 5.90 MeV and 5.93 MeV which are precisely the energies predicted from the neutron data for the $J^\pi = 9/2^+$, $11/2^-$ and $13/2^+$ rotational levels. As expected, the lower-spin members of the rotational bands are badly-resolved due to their smaller energy spacing. The fission fragment angular distribution data are shown in figure 3 for six E^* intervals corresponding to (i) : three broad intervals covering the 5.826 MeV $\leqslant E^* \leqslant$ 5.892 MeV region and (ii) : the $9/2^+$, $11/2^-$ and $13/2^+$ above mentioned resonances. At once, the experimental data indicate that one must be dealing with $K = 1/2$ states (peaking of the angular distribution at zero degree) and that J increases with E^* (increase of the W(5°)/W(85°) ratio). Hence, the observed fine structure and FFAD can probably fit the rotational band scheme proposed by Blons *et al.* [10].

Quantitative results are obtained by applying the SBF procedure [10] to both the fission probability and the fission fragment angular distribution data. The (d, pf) cross section can be written as :

$$\left[\frac{d^2\sigma}{dE^*\, d\Omega_p}\right]_{d,pf} (E_d, \theta_p, E^*) = \sum_{J\pi K\nu} \left[\frac{d^2\sigma^{J\pi}}{dE^*\, d\Omega_p}\right]_{d,p} (E_d, \theta_p, E^*) \times$$

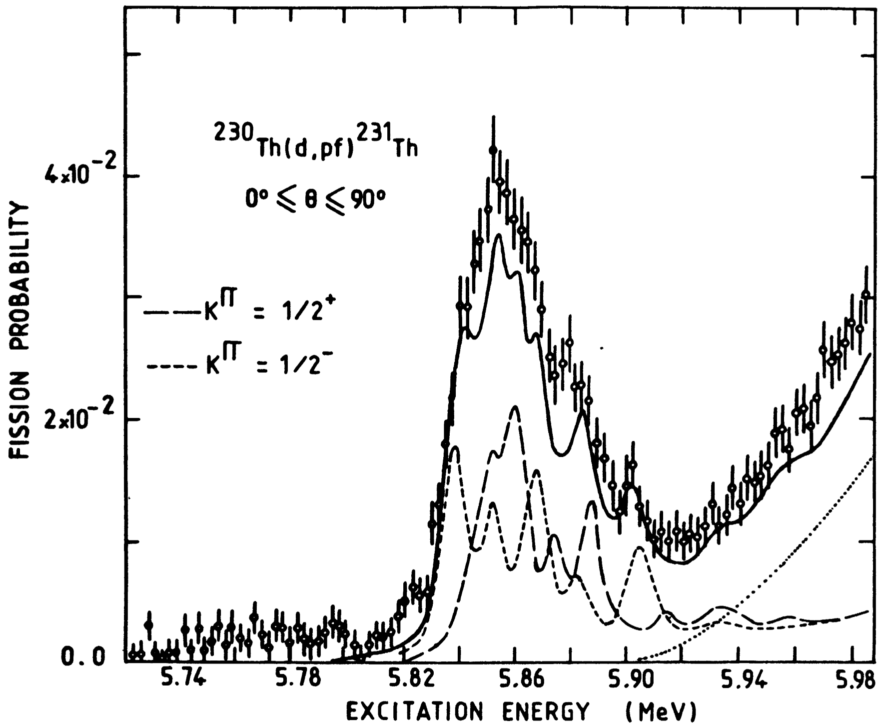
$$\times \frac{T_f^{J\pi K\nu}(E^*)}{\sum\limits_{K\nu} T_f^{J\pi K\nu}(E^*) + T_n^{J\pi}(E^*) + T_\gamma^{J\pi}(E^*)}$$

where $\left[\frac{d^2\sigma^{J\pi}}{dE^*\, d\Omega_p}\right]_{d,p} (E_d, \theta_p, E^*)$ represents the compound nucleus formation cross section by (d, p) reaction, computed with the DWBA code, DWUCK [13], using the optical potential parameters presented in table I.

The neutron and γ-ray transmission coefficients $T_n^{J\pi}(E^*)$ and $T_\gamma^{J\pi}(E^*)$ were computed as indicated in references [10, 12].

From theoretical evaluations of the THFB [14], we assume that the inner hump is much lower than the outer ones and, under these conditions, the inner barrier has no effect on $T_f^{J\pi K\nu}(E^*)$. Hence, for a specified fission exit channel, calculation of $T_f^{J\pi K\nu}(E^*)$ in the energy range of the

(a)



(b)

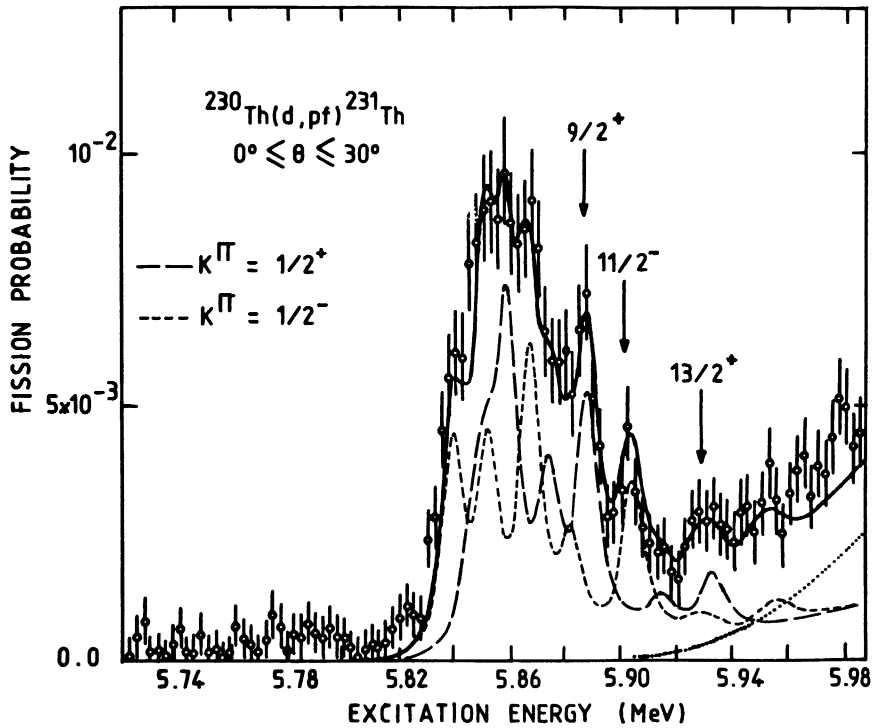


Fig. 2. — Experimental fission probabilities for 0° ⩽ θ ⩽ 90° (a) and for 0° ⩽ θ ⩽ 30° (b), compared with the SBF results (full lines). The dashed lines represent the contributions of the positive and negative parities and the dotted lines the background cross section.

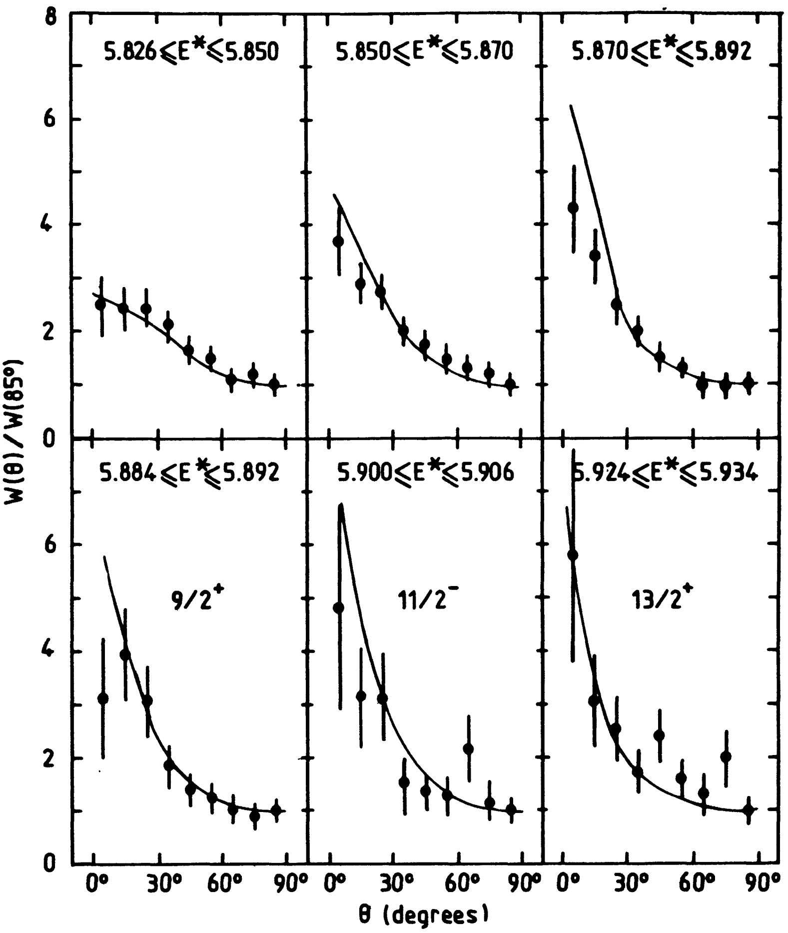


Fig. 3. — Experimental fission fragment angular distributions compared with the SBF results (full lines).

Table I. — *Values of optical potential parameters used in the calculation of the compound nucleus* (^{231}Th) *formation cross section by the* (d, p) *reaction* ($W_{vol.}$ *has been taken equal to zero*).

	V_o	$W_{surf.}$	$V_{s.o.}$	r_o	$r_{surf.}$	$r_{s.o.}$	a_o	$a_{surf.}$	$a_{s.o.}$	R_c
	(MeV)			(Fermi)						
deutons	102.3	9.2	9.0	1.1	1.33	1.1	0.82	1.02	0.82	1.3
protons	45.05	17.8	7.0	1.32	1.32	1.32	0.47	1.00	0.47	1.25

third well can be performed by using a DHFB the penetrability of which was obtained by a method devised by Cramer and Nix [15].

The heights and curvatures of the $J^{\pi} = 1/2^{-}$ and $J^{\pi} = 1/2^{+}$ barriers are taken from version C of reference [10]. These barriers are identical in shape and differ only by the inversion energy of

8 keV. Similarly, the barriers ascribed to the different J^π rotational states are supposed to be the same as the one for the $J^\pi = K^\pi = 1/2^\pi$ band head, but shifted by the appropriate rotational energy assumed to be independent of the deformation. The energies of the rotational levels, the inertia parameter, $\hbar^2/2\,\mathfrak{J}$, and the decoupling parameter, a, of the two opposite parity bands are obtained by applying a SBF procedure.

The best possible SBF to the experimental data achieved so far, is represented by full lines in figures 2a, 2b and 3. The contributions of positive and negative parities are also shown in figures 2a and 2b. The energies of the different spin components are given in table II. The background cross section above 5.90 MeV has been fitted assuming a $k = 3/2$ fission channel.

The calculated fission probability fits fairly well the experimental data for $0° \leqslant \theta \leqslant 30°$ (Fig. 2b). For $0° \leqslant \theta \leqslant 90°$, (Fig. 2a) where the fine structure features are washed out, the calculated and experimental data do not agree so well. It appears a lack of cross section in the calculated values between 5.84 and 5.89 MeV. In particular, the minimum at about 5.878 MeV coincides with a structure in the experimental data. One notices that this structure is not visible at forward angles. Such a lack of cross section could be cured by an additional contribution of high-K states. If this contribution is such that $K \geqslant 7/2$ and $K \leqslant J \leqslant 15/2$ it will certainly affect the angular distribution above $\theta = 30°$ where its maximum value occurs. Conversely, for $0° \leqslant \theta \leqslant 30°$ the high-K contribution is practically zero and it will not spoil the $K = 1/2$ angular distribution in this angular range. In fact, one could assimilate fragment detection in the forward direction as a « filter » sensitive to $K = 1/2$ only. If one considers the corresponding ^{230}Th(n, f)

Table II. — *Values of the rotational band parameters and rotation level energies.*

Parity + $\hbar^2/2\mathfrak{J}$ = (1.9 ± 0.1) keV a^+ = (0.2 ± 0.1)		Parity - $\hbar^2/2\mathfrak{J}$ = (2.0 ± 0.1) keV a^- = (-0.1± 0.1)	
J^π	E^*(keV)	J^π	E^*(keV)
$1/2^+$	5842 ± 2	$1/2^-$	5834 ± 2
$3/2^+$	5849 ± 2	$3/2^-$	5838 ± 2
$5/2^+$	5857 ± 2	$5/2^-$	5850 ± 2
$7/2^+$	5872 ± 3	$7/2^-$	5865 ± 3
$9/2^+$	5886 ± 2	$9/2^-$	5880 ± 3
$11/2^+$	5912 ± 2	$11/2^-$	5902 ± 2
$13/2^+$	5930 ± 3	$13/2^-$	5926 ± 3

data, one notes that such high K contamination must be less severe since one can hardly expect to attain a maximal value of about $K = J = 7/2$.

As for the inertia parameter $\hbar^2/2\,\Im$, the measured value of $\simeq 2$ keV confirms earlier results obtained by Blons *et al.* [7] from ^{230}Th(n, f) reaction, and differs from the value of $\simeq 3.3$ keV, obtained for fission isomers in the second well of the fission barrier [16].

One thus concludes that the present results confirm the existence of a third minimum in the fission barrier of ^{231}Th. In addition, the observed components of the rotational bands which, for the (n, f) interactions, were limited to $J = 7/2$, are now extended to $J = 13/2$.

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