Hyperdeformed fission resonances and transition states observed in $^{232}$U

L. Csige$^{1,2}$, M. Csatlós$^2$, T. Faestermann$^3$, Z. Gácsi$^2$, J. Gulyás$^2$, D. Habs$^1$, R. Hertenberger$^1$, A. Krasznahorkay$^2$, R. Lutter$^1$, H. J. Maier$^1$, P. G. Thirolf$^1$, T. Tornyi$^2$ and H.-F. Wirth$^3$

$^1$Ludwig-Maximilians Universität München, D-85748 Garching, Germany
$^2$Inst. of Nucl. Res. of the Hungarian Academy of Sci., H-4001 Debrecen, P.O. 51, Hungary
$^3$Technische Universität München, D-85748 Garching, Germany
E-mail: lorant.csige@physik.uni-muenchen.de

Abstract.

The fission probability of $^{232}$U has been measured using the $^{231}$Pa($^3$He,df) reaction with an energy resolution of 11 keV in the excitation energy region of $E^* = 4.0 - 6.4$ MeV. A number of sub-barrier fission resonances has been observed for the first time in the excitation energy range below $E^* = 4.8$ MeV and interpreted as rotational bands with a rotational parameter characteristic to a hyperdeformed nuclear shape ($\hbar^2/2\Theta = 1.96 \pm 0.11$ keV). Besides, the fine structure of the transition states could also be resolved in the fission probability and proved to have a rotational structure with a rotational parameter characteristic to a superdeformed-like nuclear configuration ($\hbar^2/2\Theta = 3.25 \pm 0.17$ keV).

1. Introduction

The observation of discrete $\gamma$ transitions between hyperdeformed (HD) nuclear states in a third minimum of the potential energy landscape with an axis ratio of $\sim 3.1$ still represents one of the greatest challenges of high-spin physics. Although a large community with 4$\pi$ gamma arrays was searching for HD states in very long experiments, no discrete HD transition have been identified so far in the studied mass region of $A\approx 120-160$ [1, 2, 3, 4].

On the other hand, the existence of HD states in the third minimum of the fission barrier in Th and U isotopes is firmly established both experimentally and theoretically [5, 6, 8, 9, 10, 11]. The identification of HD states is possible by the observation of transmission resonances in the prompt fission probability as a function of the excitation energy caused by resonant tunneling through excited states in the third minimum of the fission barrier. Moreover, the observed states could be ordered into rotational bands, with moments of inertia proving that the underlying nuclear shape of these states is indeed a HD configuration. For the identification of the rotational bands the spins and their projections onto the nuclear symmetry axis ($K$ values) can be obtained by measuring the angular distribution of the fission fragments.

Calculations of potential energy surfaces have predicted that HD minima correspond to reflection asymmetric shapes with large quadrupole and octupole deformations. Surfaces for U nuclei [6] show that hyperdeformed minima are predicted at a large quadrupole deformation of $\beta_2 = 0.9$, while the depth of the third well was estimated to be much larger than previously.
believed. In contrast to the $^{234,236}\text{U}$ isotopes, where sharp HD fission resonances have been identified in Refs. [7, 8, 11], in $^{232}\text{U}$ no clear resonance structures have been observed, but rather a broad shoulder around $E^* = 5.0 \text{ MeV}$, which was assumed to consist of resonances with $K^\pi = 0^+$ and $K^\pi = 2^+$ [12]. However, according to the theoretical calculations on the height of the fission barriers and on the depth of the third potential well [6, 5] and considering as well our previous experimental results on $^{234}\text{U}$ and $^{236}\text{U}$, the appearance of fission resonances representing HD states was expected also in the case of $^{232}\text{U}$.

The aim of the present experiment was to search for such sub-barrier fission resonances in $^{232}\text{U}$ and to assign the resonances to rotational bands via the determination of their rotational parameters ($h^2/2\Theta$).

2. Experimental setup

The experiment was carried out at the Tandem accelerator of the Maier-Leibnitz Laboratory (MLL) at Garching employing the $^{231}\text{Pa}(^3\text{He},\text{df})$ reaction with a $^3\text{He}$ beam of $E = 38.1 \text{ MeV}$ to investigate the fission probability of $^{232}\text{U}$ in the excitation energy region of $E^* = 4.0 - 6.5 \text{ MeV}$. An enriched (99%) 70 $\mu$g/cm$^2$ thick radio-active target of $^{231}\text{Pa}$ was used on a 20 $\mu$g/cm$^2$ thick carbon backing. The excitation energy of $^{232}\text{U}$ was determined from the kinetic energy of the deuteron ejectiles. The ground state Q value for the reaction is $Q=610 \text{ keV}$.

The kinetic energy of the outgoing deuterons was analyzed by a Q3D magnetic spectrograph set at $\Theta_{lab} = 35^\circ$ relative to the beam direction with a solid angle coverage of 10 msr [14]. The deuteron position in the focal plane was measured by a position-sensitive light-ion detector with individual cathode strip readout of 890 mm active length [15].

Transitions to the well-known low-lying states of $^{232}\text{U}$ [16] using the $^{231}\text{Pa}(^3\text{He},\text{d})$ reaction were applied for the energy calibration. The experimental energy resolution was deduced to be $\Delta E = 11 \text{ keV}$ (FWHM) in the energy region of interest.

Fission fragments were detected in coincidence with the outgoing deuterons by two position sensitive avalanche detectors (PSAD), allowing for a detection of the fission fragment angular correlation with respect to the recoil axis ($30^\circ < \Theta_R < 90^\circ$) and a solid angle coverage of 10% of $4\pi$.

3. Experimental results and discussion

The measured high-resolution excitation energy spectrum of $^{232}\text{U}$ is shown in Fig. 1 as a function of the excitation energy of the compound nucleus in the region of $E^* = 4.0 - 5.3 \text{ MeV}$ together with the contribution of random coincidence events (hatched histogram). Above $E^* = 4.8 \text{ MeV}$ the scale of the y-axis was multiplied by 7.5 for the better readability. A number of sharp transmission resonances have been observed for the first time in the excitation energy region of $E^* = 4.0 - 4.8 \text{ MeV}$ with widths of $\Delta E \approx 30 \text{ keV}$. The fine structure of the already previously known $K^\pi = 0^+$ and $K^\pi = 2^+$ resonances around $E^* = 5.0 \text{ MeV}$ [12] could also be resolved.

Figure 2(a) shows the fission probability spectrum between $E^* = 4.0$ and 4.8 MeV. It was obtained by dividing the deuteron spectrum containing only coincidence events from ($^3\text{He,d}$) with the deuteron spectrum from the ($^3\text{He,df}$) reaction. In order to describe the rotational structure of these resonances, overlapping rotational bands were assumed with the same moment of inertia ($\theta$) and intensity ratio for the band members as described previously by Krasznahorkay et al. [9]. The details of the fitting procedure are presented elsewhere [18]. The rotational parameter was determined to be $h^2/2\Theta = 1.96 \pm 0.11 \text{ keV}$ as a result of the $\chi^2$-analysis (Fig. 2b), which is significantly smaller than the corresponding value characterizing SD shapes ($h^2/2\Theta \approx 3.3 \text{ keV}$) as described in Refs. [9, 11]. The reduced $\chi^2$ value of the best fit was $\chi^2/F = 1.46$ (where $F$ is the number of the degrees of freedom), representing the quality of our fitting procedure.
Figure 1. Deuteron coincidence spectrum of the $^{231}$Pa$(^{3}$He,df) reaction between $E^* = 4.0–5.3$ MeV. A number of sharp fission resonances (interpreted as HD resonances below ca. 4.8 MeV and as transition states in the region of the second barrier top between 5.0 and 5.2 MeV) have been observed for the first time in this excitation energy region. The contribution of random coincidence events is indicated by the hatched spectrum. Above $E^* = 4.8$ MeV the scale of the y-axis was multiplied by 7.5 for the better readability.

Figure 2. a) Fission probability with error bars as a function of the excitation energy between $E^* = 4.0$ MeV and $E^* = 4.8$ MeV. The result of the fitting procedure assuming seven rotational bands with overlapping band members as described in the text is indicated by the continuous line. The picket fence structures indicate the positions of the rotational band members used in the fit. b) Result of the $\chi^2$-analysis. For details see Ref. [18].

One of the specific features of the HD bands in the actinide region additional to the large moment of inertia is the appearance of octupole bands. The different consequences of the octupole deformation have been reviewed in detail by Butler and Nazarewicz [13]. Experimentally the very large quadrupole and octupole moments of the HD states usually manifest themselves by the presence of alternating parity bands with very large moments of
The present experimental data could support only a very small value of the inversion parameter in the case of \(^{232}\)U which is fully consistent with the ones obtained by Blons et al. for the Th isotopes \([7]\) and with our previous experimental findings \([9, 11]\). On the other hand it has to be noted that our analysis is not sensitive to the parity of the states, so in the case of \(K \neq 0\) we cannot distinguish between quadrupole and octupole bands. However, our result on the depth of the third potential well \((E_{III} = 3.2 \pm 0.2 \text{ MeV})\) suggests that in the case of \(^{232}\)U fission proceeds rather via reconnection asymmetric shapes following the picture of Ref. \([6]\). At this moment we do not have any direct experimental information on the octupole deformation parameter \((\beta_3)\) of this nucleus.

**Figure 3.** a) Fission probability with error bars as a function of the excitation energy between \(E^* = 4.9 \text{ MeV}\) and \(E^* = 5.2 \text{ MeV}\). The result of the fitting procedure assuming two quadrupole \(K^\pi = 0^+\) bands and one \(K^\pi = 2^+\) band is indicated by the continuous line. The positions of the band members are indicated by the picket fence structures. b) Result of the \(\chi^2\)-analysis.

Going beyond the discussion presented in Ref. \([18]\), Figure 3(a) shows the fission probability spectrum between \(E^* = 4.9\) and 5.2 MeV. Considering our previous results on the fission barrier parameters of \(^{232}\)U \([18]\), the resonance structure around \(E^* = 5.0\) MeV could be a nice signature of the so-called transition states, which could be assigned to states built on top of the second barrier. According to the transition state concept introduced by Bohr \([19]\), most of the nuclear excitation energy is transformed into deformation energy when the nucleus is passing over the saddle point. Thus, at this high deformation, the compound nucleus is thermodynamically cold, resulting in a discrete spectrum of low-lying collective excitations, thus rotational bands as well. Following Bohr’s concept, the near-barrier fission proceeds through these discrete transition states, however, the rotational behavior of these states has so far not been observed.

In order to describe the rotational structure of the observed resonances, the same fitting procedure was applied as described previously for the HD bands. In agreement with the previous experimental findings of Ref. \([12]\), the resonances observed here were described by \(K^\pi = 0^+\) and \(K^\pi = 2^+\) quadrupole rotational bands up to \(J = 4^+\). Our results were deduced assuming \(K = 0, 2\) and 0 for three quadrupole rotational bands at band head energies of \(E^* = 4970(5), 4995(5)\) and 5085(6) keV, respectively. However, probably due to the limited statistics, the \(J = 3^+\) member of the \(K^\pi = 2^+\) band was not observed. The rotational parameter of the bands was determined to be \(\hbar^2/2\Theta = 3.25 \pm 0.17 \text{ keV}\) as a result of the \(\chi^2\)-analysis (Fig. 3b) in fair agreement with the value of 3.38 ± 0.10 keV published recently by Krasznahorkay et al. \([21]\).
The superdeformed character of these resonances indicates a distortion of the fission barrier at the position of the second barrier. The reduced \( \chi^2 \) value of the best fit was \( \chi^2/F = 1.21 \).

The angular distribution of the fragments with respect to the recoil axis was determined by gating on the energies of the observed rotational bands in the excitation energy region of \( E^* = 4.9 - 5.3 \) MeV. Two experimental angular distributions were generated, one for the rotational bands observed between \( E^* = 4950 \) keV and 5060 keV and another for the band having its bandhead at \( E^* = 5085 \) keV. The values were normalized to the value at 90°, as can be seen in Fig. 4. Angular data for random coincidence events were subtracted, however, errors represent only the statistical uncertainty; no estimation could be performed on the uncertainty coming from the continuous non-resonant part of the fission probability.

The measured angular distribution as a result of a preliminary reduction of the angular data was compared to the calculated one in order to support the \( K \) value assignments of the rotational bands. The calculation has been performed following the concept of Glässel et al. [20]. In Fig. 4 the different lines represent the resulting normalized angular distribution obtained from the measured resonances that were assigned to the rotational bands with their respective \( K \) values, as derived from the fitting procedure. In Fig. 4a) the dashed line represents the theoretical angular distribution for the overlapping bands at \( E^* = 4950 \) keV and 5060 keV, assuming a mixture of \( K = 0 \) and \( K = 2 \) bands. The experimental and calculated values are in nice agreement with the \( K = 0, 2 \) and \( K = 0 \) assignments for the bands at \( E^* = 4950 \) keV, 5060 keV and \( E^* = 5085 \) keV, respectively, however, the large uncertainty originating from the non-resonant fission process warns us to use these very preliminary results with precautions.

Figure 4. Comparison of theoretical and experimental fission fragment angular distributions \( W(\Theta_R)/W(90°) \) with respect to the recoil axis for the rotational bands observed at the excitation energy region of a) \( E^* = 4950 - 5060 \) keV and b) \( E^* = 5060 - 5200 \) keV. The theoretical distributions were calculated using \( K = 0, 2, 3 \) and 4 assignments as indicated in the figure as well.
4. Summary
We measured the fission probability of $^{232}$U with high resolution using the $^{231}$Pa($^3$He,df) reaction in order to search for theoretically predicted hyperdeformed fission resonances. As a result of the experiment, sharp transmission resonances have been found at the excitation energy region of $E^* = 4.0\text{–}4.8$ MeV. These resonance structures are interpreted as rotational bands with a moment of inertia characteristic to hyperdeformed nuclear states ($\hbar^2/2\Theta = 1.96 \pm 0.11$ keV). Considering the height of the previously determined inner barrier, the resonances observed near $E^* = 5.0$ MeV could be transition states exhibiting a rotational structure with a rotational parameter characteristic to superdeformed nuclear configurations ($\hbar^2/2\Theta = 3.25 \pm 0.17$ keV) which is also supported by our preliminary results on the angular data analysis.

5. Acknowledgments
The work has been supported by DFG under HA 1101/12-2 and UNG 113/129/0, the DFG Cluster of Excellence “Origin and Structure of the Universe”, and the Hungarian OTKA Foundation No. K72566.

References
[1] Lafosse D R et al. 1995 Phys. Rev. Lett. 74 5186
[2] Lafosse D R et al. 1996 Phys. Rev. C 54 1585
[3] Wilson J N et al. 1997 Phys. Rev. C 56 2502
[4] Herskind B et al. Acta Phys. Pol. B38 1421 and references therein
[5] Möller P, Nilsson S G and Sheline R K 1972 Phys. Lett. B40 329
[6] Ćwiok S et al. 1994 Phys. Lett. B322 304
[7] Blons J et al. 1988 Nucl. Phys. A477 231
[8] Krasznahorkay A et al. 1998 Phys. Rev. Lett. 80 2073
[9] Krasznahorkay A et al. 1999 Phys. Lett. B461 15
[10] Thirolf P G and Habs D. 2002 Progr. Part. Nucl. Phys. 49 325
[11] Csatlós M et al. 2005 Phys. Lett. B615 175
[12] Back B B et al. 1974 Phys. Rev. C 9 1924
[13] P.A. Butler and W. Nazarewicz 1996 Rev. Mod. Phys. 68 349
[14] Enge H A 1979 Nucl. Inst. Meth. 162 161
[15] Wirth H F 2001 Ph.D. Thesis TU Munich
[16] Browne E 2006 Nuclear Data Sheets 107 2579
[17] Sin M et al. 2006 Phys. Rev. C 74 014608
[18] Csige L et al. 2009 Phys. Rev. C 80 011301
[19] Bohr A 1956 Proc. of the 1st UN Int. Conf. on Peaceful Uses of Atomic Energy (New York) vol 1 p 151
[20] Glässel P, Rössler H and Specht H J 1976 Nucl. Phys. A256 220
[21] Krasznahorkay A et al. 2009 Proc. Int. Conf. on Nucl. Struct. and Rel. Topics (Dubna) vol 1 (JINR) p 158