Identification of a Shape Isomer in ^{235}U

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The shape isomer in ^{235}U has been searched for in a neutron-induced fission experiment on ^{234}U, which was performed at the isomer spectrometer NEPTUNE of the EC-JRC IRMM. A neutron source, with a tunable pulse frequency in the Hz to kHz range and its individually adjustable neutron pulse width in connection with an appropriate detector system turned out to be the ideal instrument to perform an isomer search, when decay half-lives above 100 μs are expected. From the delayed fission events observed for two different NEPTUNE settings and at mean incident neutron energies $E_n = 0.95$ and 1.27 MeV the isomeric fission half-life could be determined to be $T_{1/2} = (3.6 \pm 1.8)$ ms. The corresponding cross section was determined to $\sigma_{if} = (10 \pm 8)$ μb. With these results an experimental confirmation for the existence of a superdeformed shape isomer in odd-uranium isotopes is given for the first time.

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The picture of a double-humped barrier in the nuclear energy landscape has been very successful in explaining a number of phenomena observed in nuclear reaction data [1–3], where actinide nuclei are involved, see, e.g., Refs. [4–7]. The observation of an intermediate structure in subthreshold fission cross sections as well as of metastable fissioning isotopes, so-called shape isomers, found their explanation within this model. The decay mode and half-life of a shape isomer directly probes the fission barrier height and penetrability, which both are important input parameters for nuclear reaction modeling.

Since the early 1960s 33 fission isomers have been discovered in nuclei ranging from ^{236}U to ^{245}Bk [8,9]. One of the persisting problems associated with fission shape isomers is the interesting question of competing decay modes of shape isomers such as, for example, decay by γ-ray emission. This process has been observed [10] and extensively investigated in the 1980s and 1990s in ^{238}U [11,12] and ^{236}U [13]. Particularly in the latter case this decay mode dominates over isomeric fission. Recently, the predicted hyperdeformed nuclear states in the nuclear energy landscape [14,15] have been successfully introduced into cross section model calculations to evaluate neutron-induced reaction cross sections of the lightest actinide nuclei such as ^{232}Th, ^{231}Pa and ^{236}U [16]. Still there are different opinions whether or not the third minimum is shallow. Whereas in Refs. [17,18] a depth of more than 2 MeV is suggested in ^{234}U and ^{236}U, cross section calculations for ^{232}Th and ^{231}Pa isotopes still favor the old picture of a shallow well with a depth smaller than 1 MeV. It seems, that only the direct measurement of shape isomer decay properties as well as the corresponding ground state energy E_{II} provide the key to resolve the puzzle. Although general features of fission barriers and of shape isomers are fairly well understood [8], reality is obviously much more complicated. Therefore, more consistent experimental work is indispensible still after 30 years. Even more intriguing today is the lack of any shape isomer half-life data for odd-N uranium and neptunium isotopes. Only in the case of ^{239}U at least the population of the superdeformed ground state in a neutron-induced capture experiment has been observed [19,20], still neither decay nor half-life of the shape isomer was observed, yet. The nuclides ^{236}U [21,22] and ^{240}Pu [23,24] represent a few examples, where the ground state energy of the shape isomer E_{II} has been measured directly. Since fission isomer half-lives for odd-N uranium and neptunium isotopes are expected to be in the order of several hundreds of μs or even longer, the detection with commonly pulsed particle beams is extremely difficult. This is even more difficult in neutron-induced reactions, where the environmental background between pulses due to scattered neutrons is extremely disadvantageous. Together with the extremely low production cross section of shape isomers, typically smaller than 20 μb [7,25], as well as half-life predictions from different trend evaluations, ranging in some cases over 5 orders of magnitude [26–28], leaves the measurement of shape isomer decay data to a challenging venture for the experimentalist. Here, we report on the results of the shape isomer search in ^{235}U performed in a neutron-induced fission experiment on ^{234}U. In this relatively favorable case existing half-life estimates cover only 1 order of magnitude, namely, 1.2 [28], 4.8 [27], and 11 ms [26].

The experiment was performed at the NEPTUNE isomer spectrometer, which was installed recently at the Van de Graaff laboratory of the IRMM, one of the Joint Research Centers of the European Commission, located in Geel, Belgium. NEPTUNE delivers quasimonoenergetic neutron beams with pulse repetition frequencies up to 5 kHz and an adjustable pulse width and is dedicated to investigating delayed decay processes with half-lives ranging from several μs up to hundreds of ms (for details see Ref. [29]).

The fission fragments were detected with a twin Frisch-grid ionization chamber (IC) with common anode. A 1.364 mg sample of ^{234}U was placed in the center of the cathode closest to the neutron source. The contamination from ^{235}U was smaller than 9×10^{-4}. A sample with 2.38 mg of ^{235}U served as monitor for room-scattered and thermalized neutrons and was mounted on the second cathode of the IC. Both samples had a circular shape and a radius of 2.8 cm. Fast neutrons were monitored with a NE213-equivalent scintillation detector (4 inches in diameter × 1 inch thickness), employing the pulse-shape technique for neutron-gamma separation. Measurements were performed at $E_n = 0.95$ and 1.27 MeV at pulse frequencies $\nu = 100$ and 50, 100, and 150 Hz, respectively. In all settings a duty cycle of 30% was chosen. The neutron beam was produced by employing the $T(p, n)$ reaction. With an average beam current of 10 μA on a Ti:T target of initially 2 mg cm^{-2} thickness, a neutron flux of about 2.0×10^6 cm^{-2} s^{-1} was obtained at ^{234}U target position. Both incident neutron energies were chosen, since they correspond to the position of vibrational resonances in the neutron-induced fission cross section [25], and one could hope that the population of the shape isomer was enhanced due to coupling to excited states above the second minimum. This was observed in the reaction ^{238}U(n, f) around 720 eV [7,20]. The energy resolution was 300 keV (FWHM), mainly determined by the thickness of the production target and the solid angle. The frequencies were varied as mentioned above in order to optimize the half-life measurement. However, during the experiment it became soon obvious that the time period corresponding to 150 Hz was too small. Hence, only few events were measured with this setting and further analysis of them was not possible.

During two weeks of beam time, corresponding to 11.7 days actual run time, 55 delayed fission events from the ^{234}U target were identified in total. Fission-fragment pulse heights were corrected for angular-dependent energy loss based on the analysis of the pulse height of the prompt fission events as a function of the cathode signal, which is proportional to the fragments' range times the cosine of the emission angle θ [30]. Taking into account only events with $\cos\theta > 0.45$ allowed an efficient suppression of the α-particle background from the natural decay of ^{234}U. As a result, the decay time, taken relative to the end of the neutron pulse, versus the fragment pulse height is shown in Fig. 1. Also shown are the projections on the time axis (Fig. 2) as well as on the pulse-height axis (Fig. 3). Since all individual events are shown, no correction for neither ^{235}U contaminants nor scattered neutrons was performed in both figures. The double-humped pulse-height distribution, typical for the fission process, measured in the "beam off"

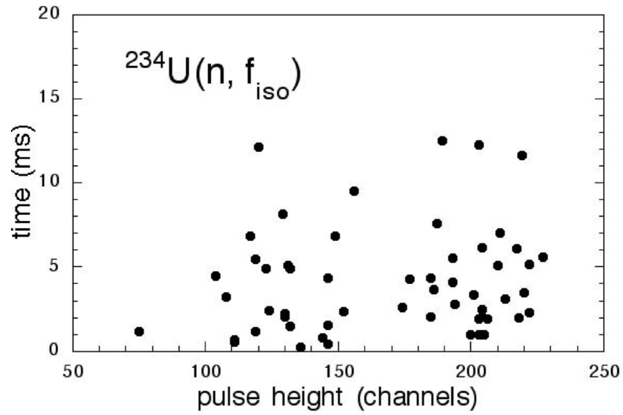

FIG. 1. Fission fragments from the decay of the shape isomer in ^{235}U*: Decay time relative to the end of the neutron pulse vs angular-dependent energy-loss corrected fission-fragment pulse height.

time interval allows us to conclude, that delayed fission events were detected. In Fig. 4 the pulse-height spectrum of fragments from prompt fission of ^{235}U* is shown for comparison. Here the contribution due to the ^{235}U admixture is negligible. The delayed ^{235}U fission events are summed in time bins of 3 ms and corrected for possible fission induced by residual background neutrons between the neutron pulses.

This background may consist of three components: first, thermalized neutrons, which have a lifetime comparable with the investigated time interval, might have induced fission in the ^{235}U admixture in the ^{234}U target; second, a time-independent background and, finally, a time-dependent background due to fast-neutron-induced fission, if the proton beam was not perfectly deflected. The first component was monitored with the ^{235}U monitor sample. For each setting, the number of "late" fission events, i.e., beam off-target, induced in the ^{235}U sample, was determined. In total, their number was 8789, which corresponds to a contribution of 4.2 (out of 55) in the ^{234}U sample. The time dependance was described by the sum of two expo-

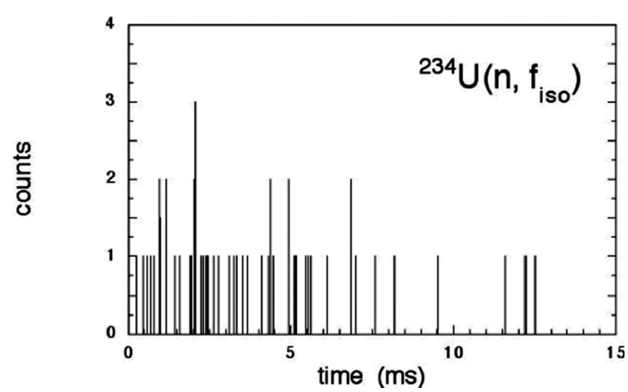

FIG. 2. Time distribution of shape isomeric fission events from ^{235}U* prior to any background subtraction.

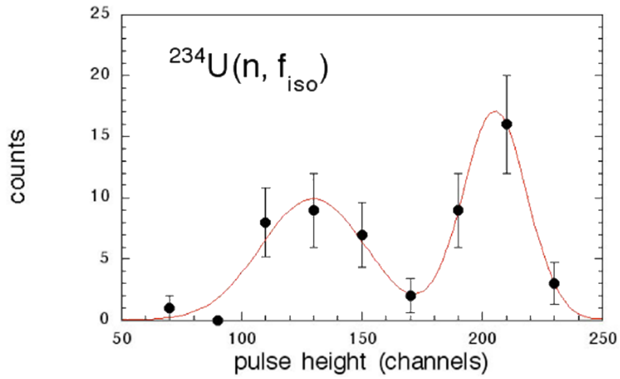

FIG. 3 (color online). Shape isomeric (delayed) fission-fragment pulse-height distribution corrected for angular-dependent energy loss prior to any background subtraction.

nentials, scaled to the ^{235}U content in the ^{234}U sample and corrected for each individual time bin. The last two components were monitored with the liquid scintillation detector. The analysis of the data for all measurements did not show any time-dependent background, and the time-independent background agrees reasonably well with the "accelerator off" condition. It was detected as proton recoil with energies larger than 3 MeV and may be attributed to cosmic rays. Assuming these particles being neutrons with an energy around 6 MeV results in at most one possible fission induced in the ^{234}U sample during one week of measurement. This, again, agrees with the fission rate obtained during an "accelerator off" run.

The final shape isomer decay curves are obtained in the following way: the observed delayed fission events are collected in time bins of 3 ms width, normalized with the time of measurement and background corrected as described above. Because of the statistical nature of decay, the detected events are not homogenously spread over a time bin. Hence, the position is usually not the middle of the bin, instead the average time of all events in the bin was chosen to represent the locations of the corresponding bins. Consequently, the error bars for the time positions are determined by the standard deviations of the time values of the events in each bin. The results are shown in Fig. 5, separately for the two incident neutron energies and fitted to exponentials. The given error bars include statistical errors as well as systematical errors with respect to the background. From both fits a weighted average half-life $T_{1/2} = (3.6 \pm 1.8)$ ms is obtained.

The shape isomer population probability was determined by dividing the integrals of the fitted exponentials with the measured prompt fission yields from $^{234}\mathrm{U} + n$. The average value is $P_{\mathrm{iso}} = (7.5 \pm 6.0) \times 10^{-6}$. With the known prompt neutron-induced fission cross section of ^{234}U, this corresponds to a production cross section of the shape isomer of (10 ± 8) μb. No statistically significant enhancement of the production cross section in the vibrational resonance at $E_n = 1.27$ MeV was observed. Combining the relative outer fission barrier height E_B and the shape isomeric ground state energy E_{II} from Ref. [7] with the measured half-life allows to calculate a penetrability $\hbar\omega_B$ according to the Hill-Wheeler approximation

$$T_{1/2}(IF) = 2.77 \times 10^{-21} \exp[2\pi(E_B - E_{II})/\hbar\omega_B]. \quad (1)$$

Using $E_B = (5.6 \pm 0.3)$ MeV and $E_{II} = (2.5 \pm 0.3)$ MeV from Ref. [7], we obtain $\hbar\omega_B = (0.47 \pm 0.06)$ MeV, which is lower than the recommended value of $\hbar\omega_B = 0.52$ MeV, also reported in Ref. [7], but still compatible within the error bars.

This result points, however, to the existence of a double-humped outer barrier in ^{235}U, well in agreement with results for ^{234}U and ^{236}U reported, e.g., in Refs. [17,18]. Other published values like $E_{II} = (2.1 \pm 0.1)$ MeV [31] and $E_B = 6$ MeV [26], respectively, are not used, since they do not represent a complete set of data. Each of them individually, however, would lead to an increase of the effective barrier height and, thus, an increase of $\hbar\omega_B$, too. In order to resolve this uncertainty, further experimen-

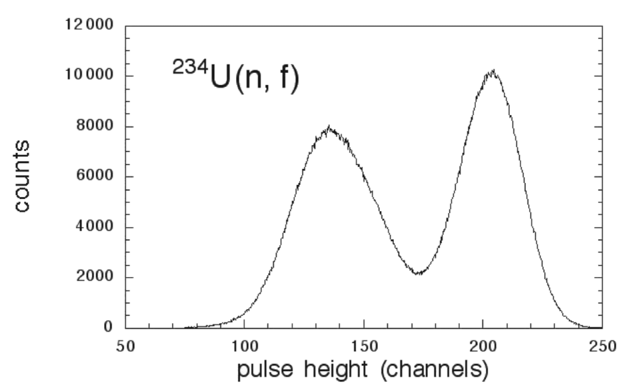

FIG. 4. Angular-dependent energy-loss corrected fission fragment pulse-height distribution from prompt fission of ^{235}U*.

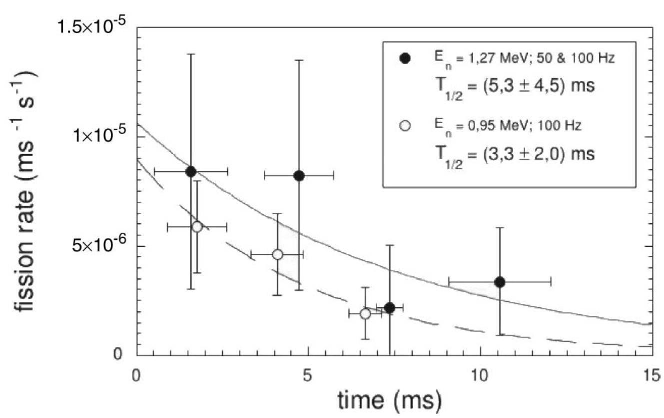

FIG. 5. Shape isomeric fission decay of ^{235}U: events are summed in time bins of 3 ms and background corrected as outlined in the text. The average half-life is $T_{1/2} = (3.6 \pm 1.8)$ ms.

tal effort is necessary; e.g., the γ decay from the second minimum back to the normal ground state should be studied.

In conclusion, we have reported on the first identification of a new superdeformed isomer in odd-uranium isotopes in the case of ^{235}U. From the measurement of neutron-induced delayed fission events, the fission half-life for the shape isomeric ground state and its population probability were determined for the first time. We reckon that the results presented in this work imply necessary and important new information for the understanding of the multihumped potential energy landscape in heavy nuclei. Actually, it is believed that the uranium and neptunium isotopes might represent a region of transition nuclides with respect to the depth of the third minimum, and amongst those ^{235}U is now the only odd-N nuclide, for which a shape isomer was identified. For these nuclides, the depth of the third minimum seems to increase compared to Th and Pa isotopes, until the third barrier drops off, starting with plutonium isotopes. We are optimistic that the presented results might trigger further spectroscopic studies as well as new theoretical work.

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