γ Decay of the Superdeformed Shape Isomer in ^{236}U

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Using the Heidelberg-Darmstadt Crystal Ball spectrometer the γ decay of the superdeformed shape isomer in ^{236}U has been investigated employing the ^{235}U(d,p) reaction at 11 MeV. The isomer was isolated by requiring that the prompt and delayed γ sum energy add up to the initial excitation energy of ^{236}U as determined from the energy of the recoiling proton. The shape isomer was found to have an excitation energy of $E(0_{II}^+)=2.75\pm0.01$ MeV and to decay by four different γ transitions in competition with its well-known fission mode. The corresponding branching ratio of γ decay to fission was determined to be $\Gamma_{F\gamma}/\Gamma_{Ff}=8\pm3$, thus resolving the long-standing problem of the missing γ-decay branch.

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An unsettled problem associated with the fissioning isomers discovered[1] almost thirty years ago in the actinide region is their γ decay, which is expected to occur in competition with the fission mode. Indeed, these isometric states have been successfully interpreted[2,3] as shape isomers located in a second minimum of the nuclear potential-energy surface with approximately twice the ground-state deformation. But so far only the decay through the outer barrier has been observed and investigated in detail,[3] while the experimental information on the electromagnetic decay back into normal deformed configurations, i.e., the tunneling through the inner barrier, is still scanty and inconclusive.

Evidence for a γ-decay branch from a fissioning isomer was first published by Russo, Petersen, and Vandenbosch,[4] in 1975, who reported on an $E2$ decay of ^{238}U to the 2^+ member of the ground-state band and possibly, on an $E1$ decay to the first excited 1^- state. They deduced a γ branching ratio with respect to the delayed fission channel of $\Gamma_{F\gamma}/\Gamma_{Ff}\approx40$ assuming that about half of the γ-decay channels have escaped observation. This ratio has to be compared with the estimate by Metag[5] of $\Gamma_{F\gamma}/\Gamma_{Ff}\approx18$, obtained by comparing the measured half-life of ^{238}U with the systematics for partial fission half-lives. The experimental results of Ref. 4 have been questioned in subsequent investigations[6,7] but are supported by a more recent experiment by Steinmayer *et al.*[8] Moreover, the monopole transition of the ^{238}U shape isomer to the ground state has been observed.[7]

For the γ decay of ^{236m}U, the only other case investigated experimentally, the semiempirical procedure of Metag[5] results in $\Gamma_{F\gamma}/\Gamma_{Ff}\approx5$, and similar values of 6 to 8 are estimated from measured delayed fission cross sections.[9-11] Despite considerable experimental efforts, however, the γ decay of ^{236m}U has not yet been observed and upper limits[12-14] of $\Gamma_{F\gamma}/\Gamma_{Ff}\approx3$–2 and ≈1.5 have been placed on the γ decay via photons of 1–2 and 2–3 MeV, respectively. In view of the results obtained for ^{238m}U, the disagreement between the experimental and expected values for the γ branching ratio of ^{246m}U is disturbing, in particular, as such a different behavior of the two uranium shape isomers is *a priori* not expected considering the comparable barrier heights and the similarity of the level schemes of both U isotopes. Clearly, further experimental effort to clarify the situation is needed. Moreover, the question of how superdeformed nuclei manage a transition from an axis ratio of 2:1 back to the ground-state value of 1.3:1 is of interest since the discovery of superdeformed bands in the mass $A\sim150$ region,[15] especially as in these cases the corresponding γ decay has not been observed either.

In the present Letter we describe the results of experiments performed on the γ decay of ^{236m}U using the Heidelberg-Darmstadt Crystal Ball[16] (CB), a 4π γ-detector system consisting of 162 individual NaI(Tl) modules. The experiments were performed using the well studied reaction ^{235}U$(d,p)^{236}$U to populate the shape isomer.[17] Metallic ^{235}U targets (90% enriched) of about 1.8- and 3-mg/cm^2 thickness were bombarded with a pulsed beam (width 25 ns, interval 280 ns) of 11 MeV supplied by the Emperor accelerator of the Max-Planck-Institut. Scattered protons were detected in a surface-barrier ring detector located at backward angles ($\bar{\theta}\approx150°$), subtending a solid angle of ≈2 sr. Particle-γ coincidences were collected for 400 h.

In order to identify γ-decaying isomers ^{236}U, the missing energy E_{missing}, which is defined by the difference of the initial excitation energy E_x in ^{236}U, determined by the proton energy, and the prompt γ sum energy $E_{\text{sum}}^{\text{prom}}$, is plotted in Fig. 1(a) against the delayed sum energy $E_{\text{sum}}^{\text{del}}$ subject to the following conditions: (i) 4.3 MeV $\le E_x\le$ 6.5 MeV, (ii) one to four γ rays are detected in the prompt time window ($\Delta t=\pm5$ ns) with respect to the proton, (iii) no detector module of the CB responded in the time window between 5 and 40 ns, and (iv) two coincident γ rays ($\Delta t=\pm5$ ns) are detected in the delayed time region between 40 and 165 ns. Isomers in ^{236}U are expected to show up on the diagonal in Fig. 1(a), as the energy which is missing in the prompt decay should appear in the delayed sum energy. Figure 1(b)

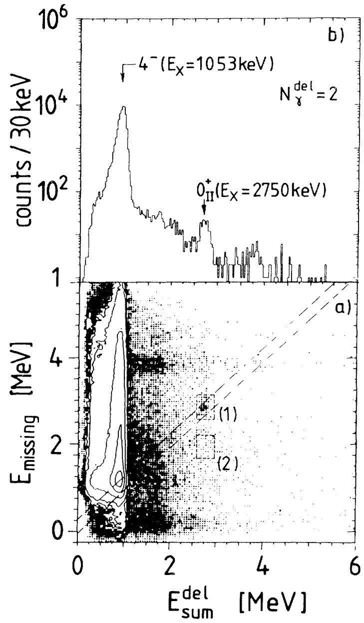

FIG. 1. (a) A two-dimensional plot of the missing energy vs the delayed sum energy for events selected to enhance transitions via isomers in ^{236}U (see text). These isomers are expected to show up along the diagonal; the projection of events within the diagonal region on the $E_{\mathrm{sum}}^{\mathrm{del}}$ axis is shown in (b). The 4^- K isomer and the 0_{II}^+ shape isomer are indicated.

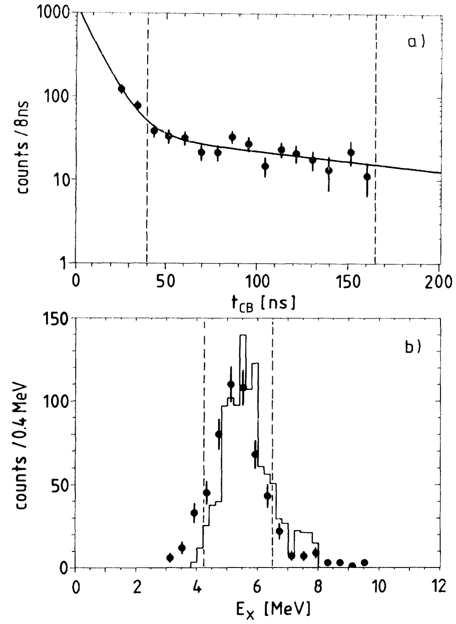

FIG. 2. (a) Time spectrum of γ rays from the decay of ^{236m}U and (b) population of excited states feeding the shape isomer, obtained by gating on events within window (1) of Fig. 1(a). The dashed regions indicate the window used for further analysis of the shape-isomer decay. The solid curves are explained in the text.

displays the projection of the events within the indicated diagonal window on the delayed sum-energy axis. Besides the well-known K isomer a further isomer at $E_{\mathrm{sum}}^{\mathrm{del}} \approx 2.7$ MeV is clearly observed, while the weak lines occurring around $E_{\mathrm{sum}}^{\mathrm{del}} \approx 1.75$ and 3.8 MeV are due to chance coincidences between a proton with prompt γ rays and delayed γ rays. The K isomer with its known γ-decay properties[18] served as an ideal test bench to quantitatively assess the procedures used in the analysis of the 2.7-MeV isomer.

The assignment of the isomer at $E_{\mathrm{sum}}^{\mathrm{del}} \approx 2.7$ MeV to the γ decay of ^{236m}U is supported by the decay curve and the distribution of initial excited states feeding the isomer. The time spectrum of the delayed γ rays leading to $F_{\mathrm{sum}}^{\mathrm{del}} \approx 2.7$ MeV is shown in Fig. 2(a). The fast component between 20 ns $\leq t_\gamma \leq$ 40 ns with $t_{1/2} = 5$ ns was also present when gated with a window (2) on the background in Fig. 1(a). The half-life of the events in the time region 40 ns $\leq t_\gamma \leq$ 165 ns was determined to be $t_{1/2} = 125 \pm 30$ ns, which agrees with the literature value of 115 ± 5 ns[11] measured for the fission decay of ^{236m}U. The population of the initial excited states feeding the isomer is shown in Fig. 2(b) (circles); it is in good agreement with the one measured by Goerlach[17] for delayed fission decay (histogram), using the same reaction and beam energy.

Figure 3(a) shows the energy spectrum of the delayed γ rays gated on the 2.7-MeV isomer [window (1) in Fig. 1(a)], while Fig. 3(b) displays the corresponding γ spectrum obtained by gating on the background window (2) in Fig. 1(a). Note, that both delayed γ rays contributing

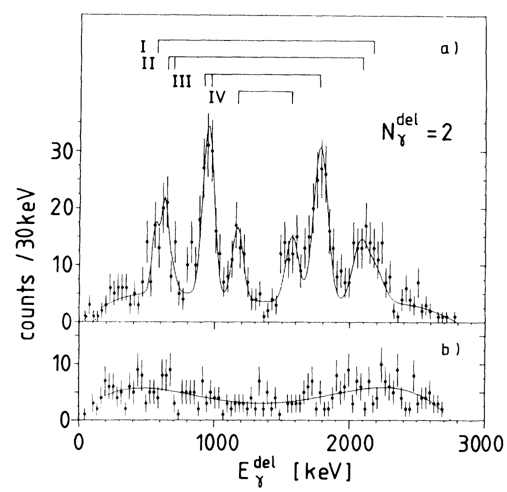

FIG. 3. (a) γ-energy spectrum for $N_\gamma^{\mathrm{del}} = 2$ events from the shape-isomer decay and (b) background decays obtained by gating on windows (1) and (2) in Fig. 1(a), respectively. The identified decay cascades are marked by I–IV.

to the spectra are detected in the neighbor-add mode. (In case two neighboring detector modules are firing simultaneously this is—for the low multiplicities discussed here—most probably caused by Compton scattering of a γ ray; consequently, the energies of both modules are added to yield the total energy of the primary γ ray, thereby increasing the full-energy efficiency.) Several lines can be observed in Fig. 3(a), which belong to four different 2γ cascades. Two of them—labeled II and III—correspond to the decay via well-known 1^- levels in the first minimum of ^{236}U at excitation energies of $E=688$ keV (II) and $E=967$ keV (III), respectively. Another cascade (IV) with γ energies $E_\gamma=1170\pm10$ keV and $E_\gamma=1580\pm11$ keV belongs to a decay via an unknown intermediate state in ^{236}U at $E=1170$ or 1580 keV, respectively. Fitting the background shown in Fig. 3(b) with a polynomial of rank 4, and using Gaussian curves for γ line shapes, a good reproduction of the measured spectrum can be obtained only if an additional cascade with γ energies of $E_\gamma=560\pm10$ keV and $E_\gamma=2190\pm30$ keV (I) is assumed. Since the level scheme is well known for excitation energies below 1 MeV and no level with $E=560$ keV is reported, it is assumed that the intermediate state has an energy of $E=2190$ keV. The four cascades amount to all but $(12\pm4)\%$ of the observed 2γ intensity.

The excitation energy of the shape isomer can be deduced from cascades II and III, by using the known excitation energies of the 1^- states. For cascades I and IV, the decay seems to proceed predominantly from the intermediate state directly into the 0^+ ground state and not into the 2^+ state, because in both cases the measured γ-ray energies add up within their statistical uncertainties to the excitation energy of the shape isomer. The deduced value of $E(0_{\rm II}^+)=2.75\pm0.01$ MeV is in agreement with the (strongly model dependent) estimate of $E\approx2.7$ MeV obtained by Back *et al.*[19] for the excitation energy of the fission isomer, but is only barely consistent with the estimate of Sood and Sarma[20] of $E=2.35$ MeV, which is based on measured average level spacings in the second minimum.

The measured $\gamma\gamma$ angular correlations for all four cascades are consistent with the assumption that the first transition of each cascade proceeds via a pure $E1$, $0^+\to1^-$ decay, supporting the 0^+ assignment expected for the shape isomer.

The γ decay of the shape isomer by cascades of one or three γ rays was also investigated. In fact, $N_\gamma^{\rm del}=3$ events are expected and observed for two reasons: (i) There is a 10% probability for Compton scattering of a γ ray into non-neighboring detectors; 30% of the observed $N_\gamma^{\rm del}=3$ intensity can thus be explained by Compton scattering of γ rays from two γ cascades. (ii) The intermediate 1_2^- state at 967 keV has a probability of 39% to decay by a two-photon cascade via the 1_1^- state rather than by a single γ ray.[18] This accounts for 50% of the observed $N_\gamma^{\rm del}=3$ events. The remaining intensity of $N_\gamma^{\rm del}=3$ events has to be attributed to further 3γ cascades depopulating the isomer, but could not be investigated in more detail because of limited statistics. These unidentified 3γ cascades contribute $\approx3\%$ to the γ decay of the fission isomer. Delayed 1γ cascades are expected in case of an $E2$ decay of the fission isomer to the 2^+ member of the ground-state band as the subsequent $2^+\to0^+$ transition is completely converted. However, the corresponding $E_\gamma=2705$ keV γ ray was not observed in the $N_\gamma^{\rm del}=1$ spectrum. An upper limit of 7% can be placed on its contribution to the γ decay.

The delayed fission decay of the shape isomer was not measured in this experiment; however, we were able to detect prompt fission events through their characteristic γ-ray signature by requiring high γ multiplicities and high γ sum energies. The branching ratio of γ decay to delayed fission $(\Gamma_{F\gamma}/\Gamma_{Ff})$ of the shape isomer was thus determined from the measured ratio of γ decays of ^{236m}U to prompt fission of ^{236}U and the known ratio of prompt to delayed fission[17] for our kinematical conditions. The resulting branching ratio amounts to $\Gamma_{F\gamma}/\Gamma_{Ff}=8\pm3$, which proves true that the main decay mode of the shape-isomer ^{236m}U is indeed the γ decay. The determined branching ratio is in agreement with various attempts[5,9–11] to understand the measured fission yields and the half-life systematics.

The decay scheme of the $0_{\rm II}^+$ shape isomer in ^{236}U deduced from the present measurement is shown in Fig. 4; if not stated otherwise γ intensities are given, which were obtained from the measured γ-cascade intensities taking into account the known internal conversion coefficients[18] in the subsequent decay of the 1_1^- and 1_2^- states. The relative accuracy of the γ branches is about $\pm20\%$.

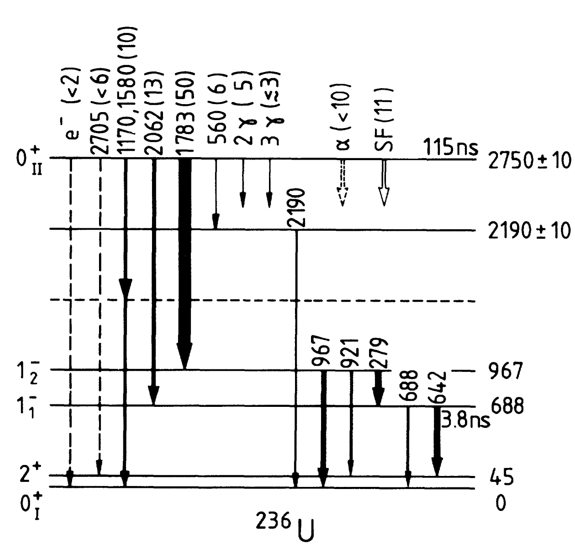

FIG. 4. Decay scheme of the superdeformed $0_{\rm II}^+$ shape isomer in ^{236}U. Transition energies are in keV and intensities are given in units of 100.

Also shown in Fig. 4 is an upper limit for the $E0$ transition to the ground state, which was deduced from the cross-section limit given in Ref. 21 and an upper limit for the α decay.[14] Note that the partial branching ratio of γ decays involving γ rays with energies $\gtrsim 2$ MeV to fission is found to be $\lesssim 1.2$, in agreement with earlier limits;[12-14] however, the intensity of the 1783-keV transition exceeds the upper (2σ) limit of Ref. 12 by about a factor of 2. The γ decay of ^{236m}U proceeds mainly via $E1$ transitions to the first and second excited 1^- state but also for the other γ branches an $E1$ character is suggested by the γ-correlation data. The corresponding $B(E1)$ values are of the order of $10^{-(10\pm0.5)}\ e^2\mathrm{fm}^2$ as is the reported $B(E1)$ value[4,8] for ^{238m}U, while the $B(E2)$ value is $<2\times10^{-6}\ e^2\mathrm{fm}^4$ in ^{236}U and $8\times10^{-6}\ e^2\mathrm{fm}^4$ for ^{238m}U (Refs. 4, 7, and 8). These values are about a factor of 10^7–10^8 smaller than for corresponding transitions between normal deformed states in the first minimum. This may be explained by the penetrability through the inner barriers, which is estimated to be of the order of 10^{-8}–10^{-9} using parameters given in Ref. 2. In a more detailed picture[2] the γ decay proceeds via tiny admixtures of normally deformed states in the wave function of the shape isomer. Within this framework the splitting of the γ strength as well as the fluctuations of the observed $B(E\lambda)$ values of up to a factor of 10 for ^{236m}U and between ^{236m}U and ^{238m}U seem to be plausible.

In summary, our detailed investigation of the γ decay of the superdeformed shape isomer in ^{236}U solves the long-standing problem of the missing γ-decay branch. Previous results on ^{238m}U are consistent with our findings but a more complete measurement of the individual γ branches of ^{238m}U seems to be desirable. The present results hopefully help to initiate also more refined theoretical approaches towards a quantitative understanding of the γ decay of superdeformed states.

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