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Spectroscopy of excited states in ^{234}U and search for a two-phonon state in ^{234}U

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Excited states of the isotope ^{234}U were studied employing the one-neutron transfer reaction ^{235}U$(d,t)^{234}$U at a beam energy of 11 MeV. The reaction channel was selected by an identification of the outgoing tritons in a ΔE-E measurement. In coincidence with the tritons, γ rays were also detected with the highly efficient MINIBALL spectrometer. Based on the analysis of $\gamma\gamma$ coincidence data, the level scheme of ^{234}U was extended. The 4^+ state at an energy of 1886.7 keV provides evidence for a possible two-phonon $\gamma\gamma$ excitation, which is based on three γ transitions from this state to the γ vibrational band. However, this state is populated with a high cross section via the one-neutron transfer reaction, which is not expected for a highly collective excitation. The situation is compared to other known cases from the rare-earth region and to the neighboring isotone ^{232}Th, which so far is the only known case for a two-phonon excitation in the actinide region.

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I. INTRODUCTION

Nuclei in the actinide region with $A \geqslant 230$ show quadrupole-deformed shapes and exhibit well-developed rotational band spectra. Below the pairing gap at 1.5–2 MeV, shape oscillations in the quadrupole and the octupole degree of freedom give rise to several low-lying rotational bands, namely, the γ band with $K = 2^+$ and octupole vibrations with $K = 0^-$, 1^-, 2^-, and 3^-. There is usually also a low-lying band with $K{=}0^+$ present in this region, which might be the β vibration or some other kind of collective excitation [1]. In addition to these one-phonon excitations, the collective nuclear model by Bohr and Mottelson [2] predicts two- and more-phonon excitations at higher energies. However, in contrast to spherical nuclei, where multi-phonon excitations are quite common, there have been only a few reports of observations of unambiguous two-phonon excitations in deformed nuclei, mostly in the region of the rare-earth isotopes [3–6]. In the actinide region, only the 4^+ state at an excitation energy of 1414 keV in ^{232}Th seems to fulfill all requirements of such a state [7,8].

The double-phonon excitation is characterized by a collective state at around twice the energy of the single-phonon state. The reduced inter-band $E2$ matrix element between the rotational bands is proportional to $\sqrt{N}$, where N is the phonon number. The transition from the double-phonon to the single-phonon state is collectively enhanced. While the direct decay from the double-phonon state into the ground-state band is forbidden. For most other candidates, which are proposed in the literature for actinide nuclei, the experimental situation is less clear. Either some experimental data are missing to prove the two-phonon character, or additional experimental findings contradict the two-phonon picture. Burke states that many of these candidates are better explained as hexadecapole vibrations [9]. Since many of the two-phonon states are expected above the pairing gap, where the density of states is higher, an unambiguous identification is very demanding. Often the multi-phonon character is probably lost completely due to mixing and fragmentation. Therefore the question of whether multi-phonon excitations exist as a common feature in deformed heavy nuclei has been the subject of research and debate over the last decades [10–12] and is not finally decided yet.

The isotope ^{234}U is a neighboring heavier even-even isotone of the already mentioned ^{232}Th. Excited states and a well-pronounced band structure are known mainly from the extensive study of the direct reaction ^{235}U$(d,t)^{234}$U and particle spectroscopy with a magnetic spectrometer done by Bjørnholm *et al.* [13] almost four decades ago. The same group also studied the β decay of ^{234}Pa to ^{234}U [14]. These decay studies have been remeasured and refined by Ardisson *et al.* [15]. Up to now, these two measurements are the main sources of data related to excited states and electromagnetic transitions in ^{234}U. Other investigations focused on the collective properties of ^{234}U by means of inelastic scattering with deuterons [16], Coulomb excitation with α particles [17], and transfer reaction with heavy ions [18].

In this work, new results were obtained by measuring the γ transitions following the one-neutron transfer reaction ^{235}U$(d,t)^{234}$U with the high-resolution 4π spectrometer MINIBALL [19]. The reaction channel is selected by identification of the light charged reaction product, and $\gamma\gamma$ coincidence data are recorded. The analysis of the coincidence data allows several extensions and corrections of the known level scheme of ^{234}U. A state at an energy of 1886.7 keV will be discussed as a candidate of a two-phonon excitation, similar to the one found in the isotone ^{232}Th. It features strong γ decays

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to the rotational band built on the γ vibration. Its excitation energy is around twice the energy of the γ vibration. Both observations are necessary for an expected double-phonon state.

II. EXPERIMENTAL SETUP AND DATA ANALYSIS

Similar to the earlier measurements of Bjørnholm *et al.* [13], the ^{235}U(d,t) reaction was exploited with a beam energy of 11 MeV to populate excited states in ^{234}U. Complementary to the spectroscopy of the outgoing tritons, we focused on the γ decays of excited states employing the high-purity germanium (HPGe) detector array MINIBALL at the Cologne tandem accelerator. The MINIBALL spectrometer [19] is based on 24 large-volume, sixfold-segmented, encapsulated HPGe detectors. The HPGe detector array consists of eight individual cluster cryostats, each containing three detectors. To achieve the highest solid-angle coverage, the eight cluster detectors were mounted at a distance of 15 cm from the target as close as possible around the target chamber. The MINIBALL triple cluster detectors are not surrounded by any Compton-shield detectors. This allows a greater flexibility for different individual configurations of the array and a high efficiency, which is required especially for experiments with low beam intensity at radioactive beam facilities. The used setup had a photopeak efficiency of around 8% at γ energies of 1.3 MeV [20]. For energy and efficiency calibration of the MINIBALL spectrometer, measurements with ^{60}Co and ^{152}Eu radioactive sources placed at target position were performed before and after the experiment.

In the target chamber, two annular Si counters (100 and 1000 μm) were placed in series under backward angles of 180° with respect to the beam direction behind the target. These two annular Si counters served as a ΔE-E telescope to identify the outgoing light particles from the transfer reaction by measuring the energy loss ΔE and the rest energy E of the particles. The beam was focused through a central hole. The detectors were shielded by a tantalum cylinder going through the central hole of both detectors and a tantalum plate at the back side of the Si stop detector against direct impact of the beam particles. At the front side of the detector telescope a thin foil of Hostaphan (40 μm) was mounted. This foil induced only a small energy loss for the light reaction products. However, the copious amount of high-energetic fission fragments generated by the deuteron-induced fission of the target nuclei was completely absorbed and stopped. The Si counters covered the angle range from 146° to 167° with respect to the beam axis. A self-supporting target foil of metallic ^{235}U with a purity of 99.7% and a thickness of 3.5 mg/cm^2 was mounted in the center of the spherical target chamber. During a 10-day-long measurement, a total of 2.6×10^6 events were recorded, which are related to the population of ^{234}U by the identification of the outgoing tritons. During the experiment, only particle-γ coincidences were recorded. The different contributions from the reaction ^{235}U$(d,p)^{236}$U, inelastic deuteron scattering, and the interesting ^{235}U$(d,t)^{234}$U reaction were unambiguously separated by gating on the two-dimensional plane of energy loss vs full energy of the outgoing protons, deuterons, and tritons. γ-ray energies were recorded within a prompt coincidence time window of ±25 ns relative to the detection of a triton. In Fig. 1, the resulting γ spectrum is shown, which is gated on the triton from the ^{235}U$(d,t)^{234}$U reaction. The high background at lower energies is due to the lack of anti-Compton shields. The γ data were sorted off-line into a $\gamma\gamma$ coincidence matrix. By setting a gate on a γ energy and an appropriate additional background gate on one axis in this matrix, an energy spectrum of all coincident γ energies and a suitable background spectrum were obtained. Background subtraction reduces considerably also the continuous background from incomplete energy detection after Compton scattering, and clean spectra are obtained. In this way, known cascades of γ transitions from excited states of ^{234}U were clearly identified and an extended decay scheme was obtained.

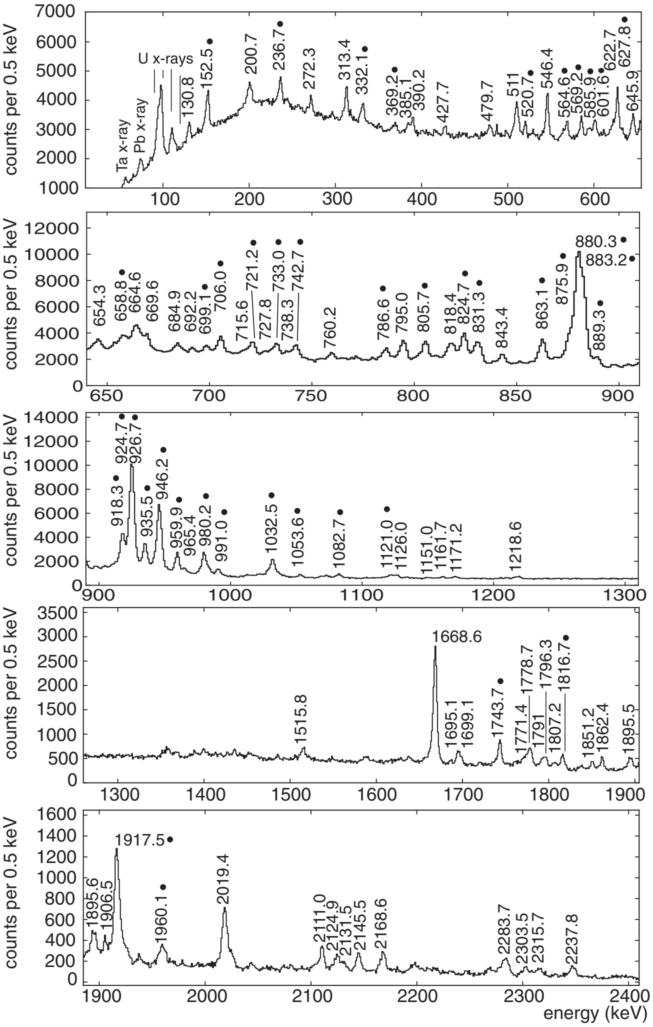

FIG. 1. γ spectrum of ^{234}U. Transition energies which are assigned to the level scheme of ^{234}U are marked by a black dot. Some of the close-lying multiplets were resolved by analyzing the $\gamma\gamma$ coincidence data.

III. LEVEL SCHEME OF ^{234}U

Figure 2 shows the level scheme as it was derived from the data of the present measurement. The widths of the lines

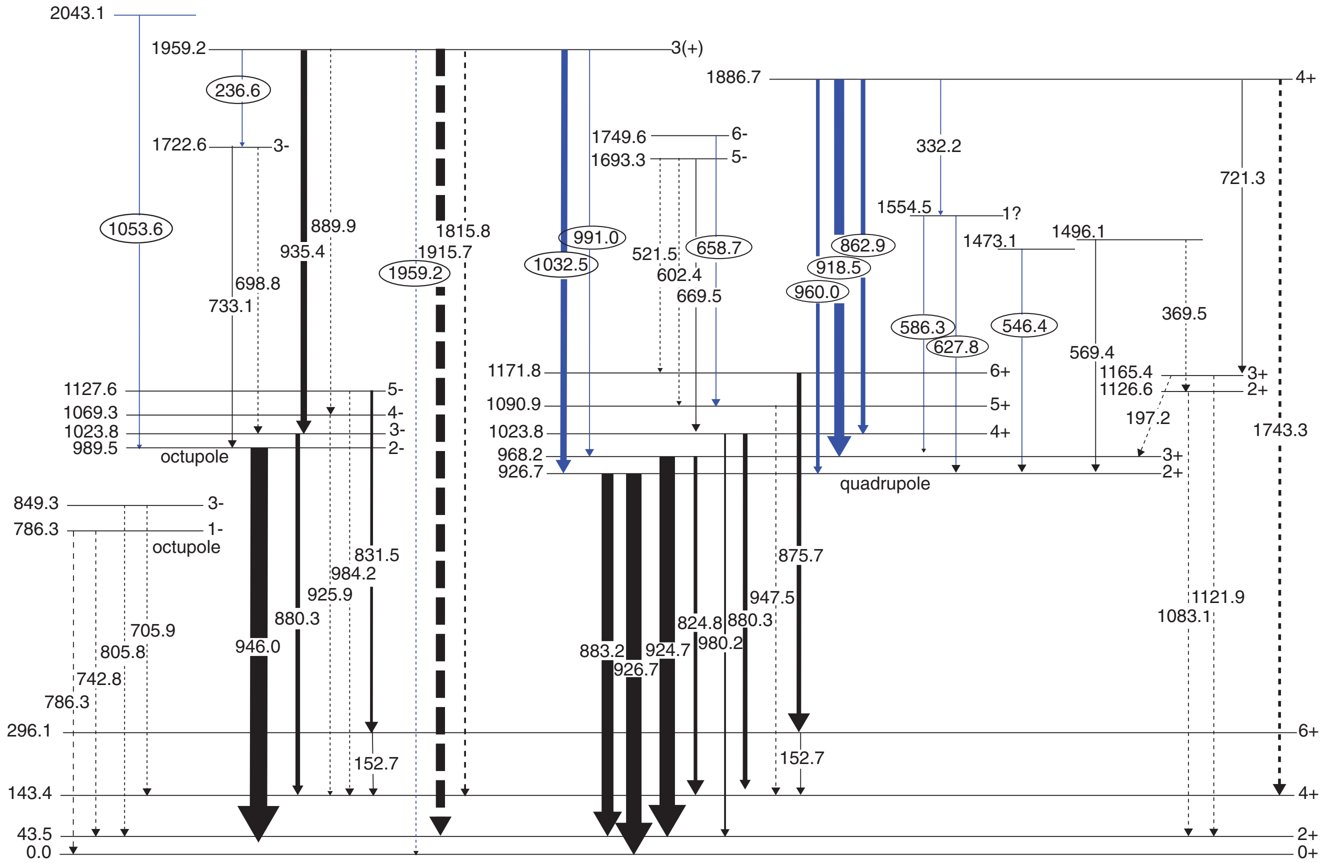

FIG. 2. (Color online) Observed γ transitions from excited states in ^{234}U. The widths of the transitions are proportional to the efficiency-corrected relative strengths of the corresponding lines in the spectrum. Transitions confirmed by coincidence data are shown as solid lines, others are shown as dashed lines. Newly assigned transitions are shown in blue (gray) and transition energies are marked by circles.

are proportional to the efficiency-corrected relative strengths of the corresponding lines in the spectrum. The strongest γ transitions are caused by the decays of the rotational states built on the γ vibration ($K^\pi = 2^+$) at an excitation energy of 926.7 keV and on the octupole vibration with $K^\pi = 2^-$ at an excitation energy of 989.5 keV. Both bands are well known from previous measurements [21]. Possible intraband transitions are not observed, since in the actinide region these low-energetic transitions decay almost completely via conversion electrons.

From previous studies of the ^{235}U(d,t) reaction [13] it is known that the octupole band is populated with a high cross section via the ^{235}U(d,t) reaction. Whereas, the rotational band built on the γ vibration is populated only weakly in the same ^{235}U(d,t) reaction [13]. The high intensity of the decays from the γ vibrational band is due to strong feeding from other states. Table I compares the relative transition strengths from the experiment with the values calculated with the Alaga rules [22]. The experimental values follow the Alaga rules quite well, except in the case of the 4^+ state. This state has within the experimental energy resolution the same energy as the 3^- state of the rotational band built on the octupole vibration with $K^\pi = 2^-$. According to earlier experiments, these two states both decay with γ energies of 880.3 and 980.2 keV. The ratio between these two transitions is expected to be 0.28 in case of the 4^+ state and 0.63 in case of the 3^- state [21]. However, the transition from the 3^- state to the 2^+ state should be very weak, since the Clebsch-Gordan coefficient for

TABLE I. Relative ratios of transition strengths for the decays of the γ band and the octupole band with $K^\pi = 2^-$ to the ground-state band. The experimental value is compared to the one calculated by the Alaga rules. The unknown multipolarities of the transitions are deduced by plausibility considerations.

	γ energies	Calc.	Expt.
Decays of the γ vibrational band ($K^\pi = 2^+$):			
$\frac{B(E2;2_\gamma^+\to 0_g^+)}{B(E2;2_\gamma^+\to 2_g^+)}$	926.7 keV 883.2 keV	0.7	0.74
$\frac{B(E2;3_\gamma^+\to 2_g^+)}{B(E2;3_\gamma^+\to 4_g^+)}$	924.7 keV 824.7 keV	2.5	2.12
$\frac{B(E2;4_\gamma^+\to 2_g^+)}{B(E2;4_\gamma^+\to 4_g^+)}$	980.2 keV 880.3 keV	0.3	0.17[a]
Decays of the octupole vibrational band with $K^\pi = 2^-$:			
$\frac{B(E3;3_o^-\to 2_g^+)}{B(E3;3_o^-\to 4_g^+)}$	980.2 keV 880.3 keV	0.0	0.17[a]
$\frac{B(E3;5_o^-\to 4_g^+)}{B(E3;5_o^-\to 6_g^+)}$	984.1 keV 831.3 keV	0.13	0.16

[a] See text for a discussion of this value.

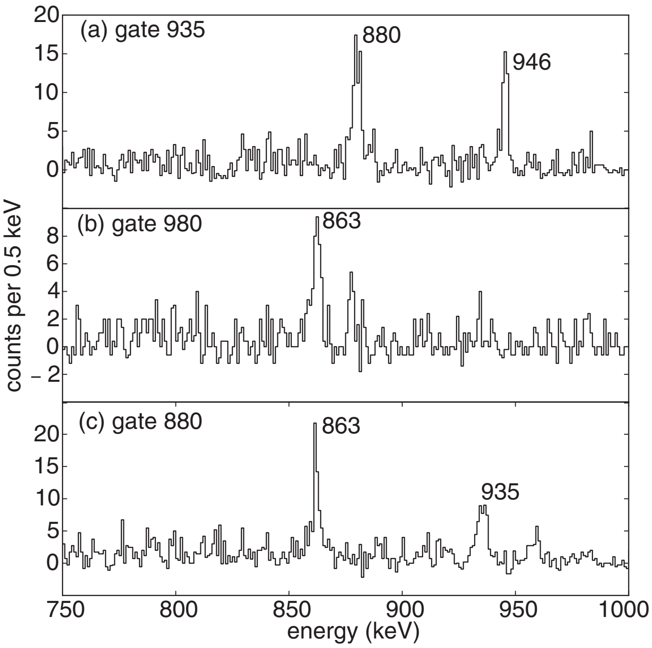

FIG. 3. Gated spectra demonstrate the decays of the states at an energy of 1023.8 keV. A gate on the energy 935.4 keV (a), feeding the 3^- state, reveals that this state decays with an energy of 880.3 keV or indirectly after a converted in-band transition via the band head of the octupole band with an energy of 946.0 keV. A γ decay with an energy of 980.2 keV cannot be confirmed to be part of the decay of the 1023.8 keV state. Additionally, this is supported by the gate on 980 keV (b), where only the transition with an energy of 862.9 keV is in coincidence. This transition populates the 4^+ state of the γ band, which has almost the same energy as the 3^- state. The last spectrum resulting from a gate on 880 keV (c) demonstrates the coincidence of this transition with both the 935.4 keV transition populating the 3^- state and the 862.9 keV transition populating the 4^+ state.

an $E3$ transition is zero. And indeed, if we assume that the strength of the line with the energy of 980.2 keV originates completely from the allowed and favored decay of the 4^+ state, whereas the strength of the other line is divided equally between the two possibilities, then the experimental value of the relative strength for the decays of the 4^+ is 0.34 and thus in good agreement with the Alaga rules. Furthermore, coincidence data agree well with this scenario. The γ rays with transition energy of 980.2 keV are not coincident with the transition with energy of 935.4 keV feeding the 3^- state. In contrast, in the case of the energy of 880.3 keV the coincidence with 935.4 keV is clearly observed. The corresponding gated spectra are shown in Fig. 3. The transition with the energy of 862.9 keV, which is also observed in these spectra, is in mutual coincidence with 880.3 and 980.2 keV and it feeds the 4^+ state.

Another low-lying rotational band in ^{234}U is built on the octupole vibration with $K^\pi = 0^-$. The band head of this band is situated at an energy of 786.3 keV. The measured intensities of the γ decays from this band are rather weak. From an earlier experiment [13] it is known that this band is not populated directly in the (d,t) reaction. Feeding transitions to this band have not been observed, since they are probably also highly converted [21].

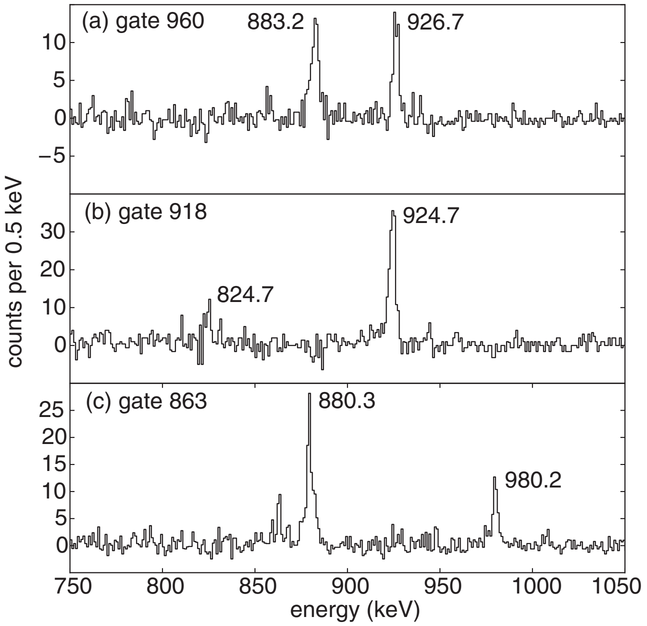

FIG. 4. Three transitions from the 4^+ state at an energy of 1886.7 keV are feeding the first states of the γ band. Spectra with gates on transition energies of 960 (a), 918 (b), and 863 (c) keV are shown. The consecutive coincident inter-band γ decays from the γ band into the ground state are identified.

A. Decays of a state at 1886.7 keV

In earlier measurements, a 4^+ state at approximately 1884.3 keV excitation energy was populated in the (d,t) reaction [13]. A more recent experiment yielded an excitation energy of the 4^+ state which has a 4 keV lower value [21]. From our level scheme, we deduce the excitation energy of this state to be 1886.7 keV, which is slightly above the first value. The 4^+ state at 1886.7 keV is established via coincidence relationships. A group of three transitions with energies of 960.0, 918.5, and 862.9 keV are measured in coincidence with strong decays from the members of the γ band. The consecutive transitions from the $2^+_\gamma \rightarrow 0^+_{\text{g.s.}}$ and the $2^+_\gamma \rightarrow 2^+_{\text{g.s.}}$ in the ground-state band are in coincidence with the 960.0 keV transition feeding the 2^+_γ state [see Fig. 4(a)]. The intermediate 3^+_γ and 4^+_γ states are observed to be in coincidence with the two pairs of transitions $3^+_\gamma \rightarrow 2^+_{\text{g.s.}}$, $3^+_\gamma \rightarrow 4^+_{\text{g.s.}}$ as well as the $4^+_\gamma \rightarrow 2^+_{\text{g.s.}}$, $4^+_\gamma \rightarrow 4^+_{\text{g.s.}}$ transitions. The corresponding gated spectra are displayed in Figs. 4(b) and 4(c). The energy of this state at 1886.7 keV is almost twice the energy of the band head of the γ vibration, and the depopulating transitions are feeding predominantly the members of the γ band. Therefore the scenario is discussed to which extent the state at 1886.7 keV excitation energy might be a candidate for the two-phonon $\gamma\gamma$ vibration.

The three transition energies of 960, 918, and 863 keV were known from previous experiments in ^{234}U [21]. However, the old findings do not agree with the new results and with the observation of these coincidences. Based on the observed coincidences, these lines are assigned to the decays of the 1886.7 keV state.

TABLE II. Relative ratios of transition strengths for the decays of the 4^+ state at 1886 keV and the $3^{(+)}$ state at 1959 keV. The experimental values are compared to results obtained by applying the Alaga rules. The unmeasured multi-polarities of the transitions are assumed to be collective $E2$ transitions.

	γ energies	Calc.	Expt.
Decays of the $K^\pi = 4^+$ state at 1886 keV:			
$\frac{B(E2;4^+\rightarrow 2^+_\gamma)}{B(E2;4^+\rightarrow 3^+_\gamma)}$	959.9 keV 918.3 keV	1.8	0.34
$\frac{B(E2;4^+\rightarrow 2^+_\gamma)}{B(E2;4^+\rightarrow 4^+_\gamma)}$	959.9 keV 863.1 keV	5.1	0.49
$\frac{B(E2;4^+\rightarrow 3^+_\gamma)}{B(E2;4^+\rightarrow 4^+_\gamma)}$	918.3 keV 863.1 keV	2.9	1.5
Decays of the $K^\pi = 3^{(+)}$ state at 1959 keV:			
$\frac{B(E2;3^{(+)}\rightarrow 2^+_\gamma)}{B(E2;3^{(+)}\rightarrow 3^+_\gamma)}$	1032.5 keV 991.0 keV	0.9	1.9
$\frac{B(E1;3^{(+)}\rightarrow 3^-_o)}{B(E1;3^{(+)}\rightarrow 4^-_o)}$	935.5 keV 889.3 keV	7.0	3.5

The relative ratios of transition strengths for these three strong transitions from the 4^+ state to the γ band are compared with the values from the Alaga rules in Table II. The deviations of the results are quite large, which may be explained either by a wrong spin assignment or by strong Coriolis mixing. Indeed, Bjørnholm *et al.* [13] discussed a strong Coriolis mixing between this state with a configuration of 1/2[501] + 7/2[473] and the 3^+ state at an energy of 1959.2 keV. According to [13], the latter state has the configuration 1/2[501]−7/2[473].

In addition to the three strong transitions, there are candidates for other decays of this 4^+ state. A triple coincidence indicates a decay via an intermediate state at 1554.5 keV, decaying farther into the γ band. The intermediate state can be identified with a known state with a spin of $J = 1$ and unknown parity [16].

From the literature, three decay transitions of the 4^+ state were expected [21]. The strongest of these transitions should have an energy of 1590.6 keV. However, there is only a very weak line with an energy differing by around 2 keV from the expected value in our spectrum. For the second transition with the energy of 1743.3 keV, there is a clear candidate in the singles spectrum. The third transition is expected to have an energy of 721.5 keV, populating an intermediate 3^+ state at an excitation energy of 1165.4 keV. This intermediate state decays with a transition energy of 197.2 keV to the 3^+ state of the rotational band built on the γ vibration. The low-energetic transition to this state cannot be observed directly due to internal conversion, but the coincidence of the transition with the energy of 721.3 keV with the decays of this state is confirmed. Moreover, the intermediate 3^+ state should decay via two transitions directly to the ground-state band. From these two transitions, the one with an energy of 1121.9 keV is observed in the singles spectra. However, it is too weak for a clear confirmation of the expected coincidence with the populating transition. The second one, expected at 1022 keV, should have comparable strength but is not observed in our data.

In summary, all known single-phonon vibrations are observed and the measured transition strengths are in good agreement with the expected values from the Alaga rule. Although the decays of the 3^- state of the octupole band and the 4^+ state of the γ band cannot be resolved, the thorough analysis of the coincidence data yields that the 3^- state is not decaying to the 2^+ of the ground band.

B. Decays of a state at 1959.2 keV

In the coincidence spectra of the decays of the γ band, there is another pair of strong lines at energies of 991.0 and 1032.5 keV. These transitions are proposed to be decays of a state at an excitation energy of 1959.2 keV. This state should have spin 3; a parity assignment does not exist [15,21]. These transition energies have been previously observed in ^{234}U, but have been interpreted differently. The transition with 989.5 keV was assigned to be emitted from a 3,4^+ state at 1916.26 keV, feeding into the 926.72 keV state of the γ band. A 5^+ state at 2101.43 keV was placed so as to decay with 1032.8 keV into the 4^- state at 1069.28 keV of the octupole band.

In β-decay studies, there has been the observation of a 3^- state at 1958.8 keV [15], while in the (d, t) measurement a 3^+ state at an energy of 1955.8 keV was observed and assigned to the Nilsson configuration 1/2[501]−7/2[743]. The known decay of this state via two transitions to the $K^\pi = 2^-$ octupole band is confirmed by the present measurement via coincidence relationships. Moreover, a triple coincidence is observed with an intermediate 3^- state at an energy of 1722.6 keV and two transitions into the octupole vibrational band. There is weak evidence for the direct decay to the ground-state band which was reported to exist in Ref. [21]. However, those lines correspond to a slightly higher excitation energy of 1960.2 keV. This is in contrast to all other transition energies, which agree very well with an excitation energy of 1959.2 keV. Other decays, listed in the literature [21], are not confirmed by our data. The relative ratios of transition strengths are shown in Table II for the two new strong transitions from the state at 1959.2 keV to the γ band members. Like for the decays of the 4^+ state at 1886 keV, considerable deviations from the Alaga rules are obvious and may be explained by Coriolis mixing between the two states.

The negative parity of the state at 1958.8 keV is deduced from the log ft values observed in the β-decay measurement [15]. Strong transitions from a state at 1959.2 keV into the γ band are measured in this experiment which favors a positive parity. Therefore a clear parity assignment can only be based on a future, specific measurement, which should also exclude the option of a close-lying doublet of neighboring states.

C. New assignments

Several new γ transitions can be placed into the level scheme of ^{234}U. A coincidence between a γ ray with an energy of 946.0 keV and an unknown γ line at 1053.6 keV is observed. An explanation for this finding is a state with an excitation energy of 2043.1 keV, depopulating via a 1053.6-keV transition directly into the 2^- state at 946.0 keV. This state is probably the same as the one observed at an excitation energy of 2038.6 keV in the earlier (d, t) measurement [13]. There is a γ energy of 546.4 keV observed in coincidence with the decays of the 2^+ state at 926.7 keV. The new γ

TABLE III. Overview of γ energies (in keV) that have been interpreted differently in this work than in the Nuclear Data Sheets [21]. The new assignments are based on $\gamma\gamma$ coincidence data.

γ energy	Initial state(s) from [21]	Initial state from this work
332.2	2068.8	1886.7
569.3	1496.1/1537.3	1554.5
586.3	1927.5	1554.5
627.8	1125.3	1554.5
862.9	1811.6	1886.7
918.4	1214.7	1886.7
960.0	1809.7/1811.6	1886.7
980.3	1023.8 ($4^+/3^-$)	1023.8 (only 4^+)
1032.5	2101.4	1959.2
1959.2	2101.4	1959.2

decay is in perfect agreement with the decay of a known state at 1473.1 keV. The coincidence between a γ energy of 569.4 keV and the decays of the 2^+ state at an energy of 926.7 keV confirms the strongest known decay of a state at an energy of 1496.1 keV [15]. For the next weaker transition with an energy of 369.5 keV, there is a candidate in the singles spectrum. However, statistics are not sufficient to be confirmed by coincidence information. The known decays of the 5^- state at an excitation energy of 1693.3 keV to states of the γ band [15] are also confirmed based on coincidences. And there is also a hint of a transition between the 6^- state at 1749.6 keV and the 5^+ member of the γ band.

These reassignments result in the reinterpretation of several strong γ transitions, which had been misplaced in the level scheme. Table III lists the differences between our and previous assignments. In addition, five transitions are newly assigned to the level scheme and one level is proposed, which may be identical to a previously known level with a similar energy.

IV. TWO-PHONON STATE

The collective nuclear model by Bohr and Mottelson [2] predicts the existence of multi-phonon states built on all one-phonon states. In the harmonic case, the excitation energies of those multi-phonon states are given by the sum of the energies of the involved one-phonon states. Transitions from one multi-phonon state to the next multi-phonon state are collectively enhanced, whereas all transitions creating or destroying more than one phonon are not allowed in that picture and therefore should be weak. The two-phonon states, resulting from the coupling of two γ phonons, depend on the final angular momentum with quantum numbers $K = 4^+$ and $K = 0^+$. These states are expected to feature a strong transition strength to the rotational band of the γ vibration. These transitions have to have a relatively pure $E2$ character and the value of $B(E2, 4^+ \rightarrow 2^+_\gamma)$ should be high. In spherical nuclei, the observation of such multi-phonon states is common, whereas their existence in deformed nuclei has been very rare and controversial [9–12].

In the actinide region, the only clear identification of such a state is known from ^{232}Th, which is a direct isotone of ^{234}U. There, the two-phonon $\gamma\gamma$ state with $K = 4^+$ was found in a Coulomb excitation experiment [7] at an energy of 1414 keV, which is 1.8 times the energy of the γ vibration and well below the pairing gap. Moreover, the lifetime and the absolute transition strength were determined, in order to prove the two-phonon character [8]. A later experiment questioned the spin and parity assignment for this state [23]. However, a few years later a reinvestigation of that matter confirmed again the two-phonon interpretation [24].

The level scheme of the isotone ^{234}U shows three vibrational states below the pairing gap, the γ vibration with an excitation energy of 926.7 keV, the octupole vibrations with $K^\pi = 0^-$ at an energy of 786.3 keV and with $K^\pi = 2^-$ at 989.5 keV. However, no multi-phonon states have been identified. Since collective features usually vary smoothly between neighboring nuclei, the existence of the two-phonon $\gamma\gamma$ state should be expected. The new $\gamma\gamma$ coincidence data establish three strong γ transitions from the 4^+ state at an energy of 1886.7 keV to the γ vibrational band. The excitation energy of this state is 2.035 times the energy of the γ vibration at an energy of 926.7 keV. Therefore, the state at 1886.7 keV is regarded to be a good candidate for the two-phonon $\gamma\gamma$ state with $K = 4^+$. However, the $E2$ character of the transitions to the γ band and the absolute value of $B(E2, 4^+ \rightarrow 2^+_\gamma)$ demonstrating the collectivity of the transitions could not be measured in this experiment. Thus a follow-up experiment is needed to clarify those points.

To first order, in a direct transfer reaction, nucleons are interchanged between target and beam nuclei without changing the configuration of the other nucleons. Therefore, transfer reactions are not very selective to collective states, but populate quasiparticle states. Since the odd neutron of the target ^{235}U has the Nilsson configuration 7/2[743], all states in ^{234}U observed in the one-neutron transfer reaction are two-quasiparticle states with one neutron in that orbital. The population of collective states is nevertheless possible, if they contain a sufficient admixture of accessible quasiparticle states. According to the microscopic-macroscopic model, the one-phonon states consist of a superposition of several two-quasiparticle states. In the earlier transfer experiments of Bjørnholm *et al.* [13], the members of the γ vibrational band were populated in the reaction ^{235}U(d, t) rather weakly via the configuration 7/2[743]−3/2[761]. In the reaction ^{233}U(d, p), the same states are populated much more strongly via the configuration 5/2[633]−1/2[631] [13]. This is consistent with the calculations of Soloviev and Siklos [25]. According to them, the main microscopic components of the γ vibration are 5/2[633]−1/2[631] with 40%, 3/2[631] + 1/2[631] with 16%, and 7/2[743]−3/2[761] with 10%. Since each phonon consists of a superposition of two-quasiparticle states, the two-phonon states should contain four quasiparticles. A population of such states is therefore not expected in the one-neutron transfer reaction. The 4^+ state at an energy of 1886.7 keV, however, is populated in the ^{235}U(d, t) with a high cross section, whereas it is not observed in the ^{233}U(d, p) at all [13]. Based on the experimental evidence and the calculations of Soloviev and Siklos, Bjørnholm *et al.* concluded that the 4^+ state at an energy of 1886.7 keV is as a pure two-quasiparticle state with the configurations of 7/2[743] + 1/2[501] [13]. Other experiments, more suitable to studying collective properties, did not cover the 4^+ state, either because their maximal excitation energy was not high enough [16,17] or they focused entirely on the ground-state band [18].

The situation found in the isotope ^{234}U is very similar to other cases, where a candidate for a two-phonon state is discussed because of strong transitions to the γ vibration. However, contradicting and incomplete experimental data do not allow an unambiguous assignment in our case. Burke [9] presented an overview of such cases in the rare-earth region. Interestingly, large cross sections in transfer reactions are quite common and indicate that these 4^+ states have large admixtures of two-quasiparticle configurations. In many cases, the states in question are identified with the strongest peaks in the corresponding spectrum. It is pointed out that all of those cases can be alternatively described as hexadecapole vibrations [9]. This would be consistent with the observed large cross sections in the transfer reactions. This interpretation is supported by calculations of Soloviev and co-workers [26], using the quasiparticle phonon nuclear model (QPNM) and by a corresponding description in terms of the interacting boson model including *g* bosons (*sdg* IBM) by Devi and Kota [27]. It is claimed in [9] that the hexadecapole interpretation would also satisfactorily explain the observed large $B(E2)$ values between the states in question and the γ vibration. Therefore, collective transitions with strong $B(E2)$ values are considered as insufficient experimental criteria for the two-phonon $\gamma\gamma$ state.

Recently, new experimental results on ^{186}Os and ^{188}Os were published. Here, the 4^+ states in question are interpreted as composition of a dominant single-hexadecapole component and a smaller, yet important $\gamma\gamma$ admixture [28]. This interpretation is supported by new calculations within the quasiparticle-phonon model [29].

From the available data, it seems that the 4^+ state at an energy of 1886.7 keV in ^{234}U can be a candidate for a two-phonon state as well as for a hexadecapole vibration. Here, new theoretical calculations would be helpful to elucidate these two options. Future experiments should examine the $E2$ character of the transitions and determine the absolute $B(E2)$ values. In this context, it seems worthwhile to investigate the state at 1959.2 keV, which is also strongly populated in the (d, t) reaction and shows strong transitions to the octupole and the γ vibrational band. In the earlier transfer-reaction studies [13], this state was interpreted as a 3^+ state built from two quasiparticles with the same Nilsson orbitals as the 4^+ state. Indications for strong Coriolis mixing between these two states were found. This is the most obvious reason for the observed branching ratios and the discrepancy with the calculated values based on the Alaga rules. In the neighboring isotope ^{232}Th, the two-phonon interpretation is established best and uniquely in the actinide region due to the long-lived ^{232}Th target material. However, no data from transfer reactions are available due to the short-lived neighbors, and the population of the two-phonon state via a transfer reaction cannot be probed in this case.

V. CONCLUSION

In summary, the new in-beam γ-ray spectroscopy experiment enabled the derivation of an extended level scheme of ^{234}U. In comparison with previous investigations in the actinide region, the high efficiency of the modern MINIBALL γ-ray spectrometer allowed the analysis of $\gamma\gamma$ coincidence data, which was not feasible in a previous γ-ray measurement by Ardisson *et al.* [15]. Strong γ transitions between a 4^+ state at approximately twice the energy of the γ vibrational and the γ vibrational band were found, a result which can be compared to the neighboring isotone ^{232}Th. In ^{232}Th, the state in question is a two-phonon $\gamma\gamma$ state based on lifetime measurements and absolute $B(E2)$ values [8]. In ^{234}U, the lifetime of the state and absolute $B(E2)$ values of the interesting transitions are not known. The strong population of this 4^+ state via the one-neutron transfer reaction ^{235}U(d, t) contradicts the two-phonon state interpretation. The situation is similar to cases in the rare-earth region discussed by Burke [9]. Here, arguments against the two-phonon interpretation are given and an alternative description by a hexadecapole vibration is suggested. The quest for a two-phonon state in ^{234}U cannot be decided based on the existing data.

To identify the collective character of the discussed candidate of the two-phonon state, complementary experiments in ^{232}Th and ^{234}U are of highest interest. In ^{234}U the collective properties should be probed via a Coulomb excitation experiment preferably in an experimental configuration which allows also the spin determination via angular correlation coefficients. In ^{232}Th the detection of the two-phonon state after a comparable light particle transfer reaction would be very helpful and may elucidate the open question to which extent the collective state can be populated in this way. However, the lack of suited targets close by ^{232}Th causes an insurmountable difficulty. Therefore future experimental and theoretical investigations in ^{234}U are suggested to clarify these open questions.

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