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On the two first excited $K = 0$ bands in ^{238}U and ^{240}Pu

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Abstract

E0-enhanced spectra of conversion electrons taken after (α,α') and (α,2n) reactions and after projectile Coulomb excitation improve our knowledge about first and second excited bands in ^{238}U and ^{240}Pu built on 0_2^+ and 0_3^+ states which lie anomalously close together. The two bands are of different structure.

The available knowledge about excited $K = 0$ bands in the heaviest nuclear systems stems from a limited number of experiments on accessible actinide nuclei. Of the known cases some exhibit unexpectedly close lying 0_2^+ and 0_3^+ states which add considerable complexity to an overall theoretical description. In ^{238}U, one of the otherwise best known actinide isotopes, the two states appear at 926 keV and, as we report, at 997 keV. A shade of doubt on the energy and J^π assignment to the latter state is removed by the E0 observations described below. The state at 926 keV has been interpreted by Chang et al. [1] as a γ-vibrational two-phonon bandhead, the one at 997 keV as "β vibrational". In ^{240}Pu the two states appear at 861 and 1089 keV.

We observed K electrons from E0-admixed $J \to J_g$ interband transitions with $\Delta K = \Delta\pi = 0$ and with large K-conversion coefficients: α_K(E0+E2) being of order one (but never well known because of mostly unknown γ-ray intensities [2]), whereas the typical conversion for stretched E2's is smaller by two orders of magnitude. It is because of the large E0 admixtures that electron spectra of some $J \to J_g$ transitions reveal interesting structure effects.

As compared to groundstate rotational bands (GSRBs), the level spacing in excited $K^\pi = 0^+$ bands is increasing less with spin J due to centrifugal stretching. Therefore the subsequent energies of the $0 \to 0_g$, $2 \to 2_g$, $4 \to 4_g$, ... transitions decrease usually in a regular way, leading to line separations of ten keV or more and resulting in nicely separated peaks in electron spectra recorded with Si(Li) detectors as in the present study. However, a so far unique close clustering of transitions has been reported by

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Venema et al. [2] for ^{234}U. We returned to this case with ^{232}Th targets to obtain reference spectra for our $(\alpha,\alpha')^{238}$U and $(\alpha,2n)^{240}$Pu measurements with ^{238}U targets. The spectrum with Th-targets presents the "fingerprint" of a nearly degenerate cluster of K-lines implying that in that case the initial and final bands have nearly identical moments of inertia [2]. As described hereafter, a similar fingerprint identifies a β band in ^{240}Pu as a second case of nearly identical bands. In a forthcoming paper we intend to discuss this phenomenon in a broader systematic way. The in-beam spectroscopy of conversion electrons has been performed at KVI [3], and at GSI [4] with a novel approach for handling large Doppler broadening.

The KVI studies used 25–40 MeV α beams, ^{nat}U targets of 0.6 to 1 mg/cm^2, and the less fissile ^{nat}Th targets for obtaining reference spectra. Because of the increasing β^- radiation from accumulating fission fragments, the spectra aiming at premium resolution and lowest background were always collected during the first cyclotron shifts (8-hr periods) with relatively low (<5 nA) beam currents and, consequently, poor statistical accuracy. In following shifts the count rates were increased at the expense of energy resolution and low background. The intrinsic detector resolution of 2.5 keV FWHM at 1 MeV has been approached with the Th spectrum. For the U targets the much higher induced electron background from prompt and delayed fission worsened the resolution to about 3.5 keV.

Coulomb excitation of 6 MeV/A ^{238}U projectiles by a ^{181}Ta target has been studied by electron spectroscopy at GSI with correction for large Doppler broadenings implicit to the use of fast heavy projectiles. Without precautions the Doppler effects would erode the energy resolution of the electron spectra taken after reactions with the heavy and fast moving projectiles. However, by using a novel arrangement [4], the loss of resolution could be compensated to a large extend by applying event-by-event kinematic corrections. In this arrangement four Mini-Orange spectrometers faced the target at backward angles (155°), one of them being dedicated to the present spectroscopy at energy settings around 1 MeV. The conversion electrons were measured in coincidence with scattered U-projectiles and recoiling target nuclei flying in forward direction. This allowed off-line kinematic corrections which accomplished a restoration of the resolution to 9.5 keV FWHM at 1 MeV (7 keV caused by residual Doppler effects).

Besides the K-lines of primary importance, the spectra of Fig. 1 do also show the weaker L and on close inspection M and N conversions at known positions and with known relative intensities. Owing to its low background the Th-reference spectrum shows also smaller peaks above the background level. When not L, M or N lines most of them could be identified as K-converted E2 transitions from relatively intense ^{238}U γ-bands falling outside the scope of this letter. The transmission curves of the Mini-Orange spectrometers (dashed curves in Fig. 1) have been estimated [3,4] with aid of standard sources (^{137}Cs,^{207}Bi; β-continuum of ^{90}Sr,Y). In all experiments the Mini-Orange devices [3] protected the Si(Li) detectors against the overwhelming amount of low energy δ rays, especially in the Coulex case of Fig. 1c). As shown by the inset of this figure, a fraction of the δ rays are still passing the Mini-Orange filter below its main threshold. The background in spectrum 1b) is mainly due to the β-ray continua from fission fragments. Time differentiation with respect to the beam pulses separated the prompt ($\approx$ 6 ns) E0 signals from random fission and n-induced background. Spectra of γ-ray singles were always recorded simultaneously, but did not show the anyhow weak $J \rightarrow J_g$ or $J \rightarrow J_g \pm 2$ β-band transitions. The γ radiation from GSRB and γ bands was of such high intensity that the Ge detectors had to be placed at large distances, making coincidence recordings impractical.

The spectrum with Th (Fig. 1a) and the spectrum with U (Fig. 1b) have been accumulated consecutively. Both show the same (α,α') and $(\alpha,2n)$ patterns but with poorer statistics and higher fission background (note the suppressed base line) when using the U targets. The (α,α') reactions lead to dominant K_{22} lines from $2 \rightarrow 2_g$ transitions: in spectrum 1a) K_{66} is not and K_{44} is hardly visible, but these lines appeared both in (not shown) spectra taken at 40 MeV. The $(\alpha,2n)$ reactions populate more of the higher-spin states of the $K = 0$ bands. The K_{00} line is always weak with respect to K_{22}. Energy wise the K_{00} is resolved in the case of ^{232}Th, and can be recognized on close inspection as a foot added to the K_{22} line in the case of ^{234}U and ^{238}U, but is overshadowed by the known $L_{22}(^{238}$U) line in case of ^{240}Pu. From the sequence set by ^{232}Th, ^{234}U and ^{238}U we assume that the relative $K_{00}(^{240}$Pu) intensity is equally small. However, a

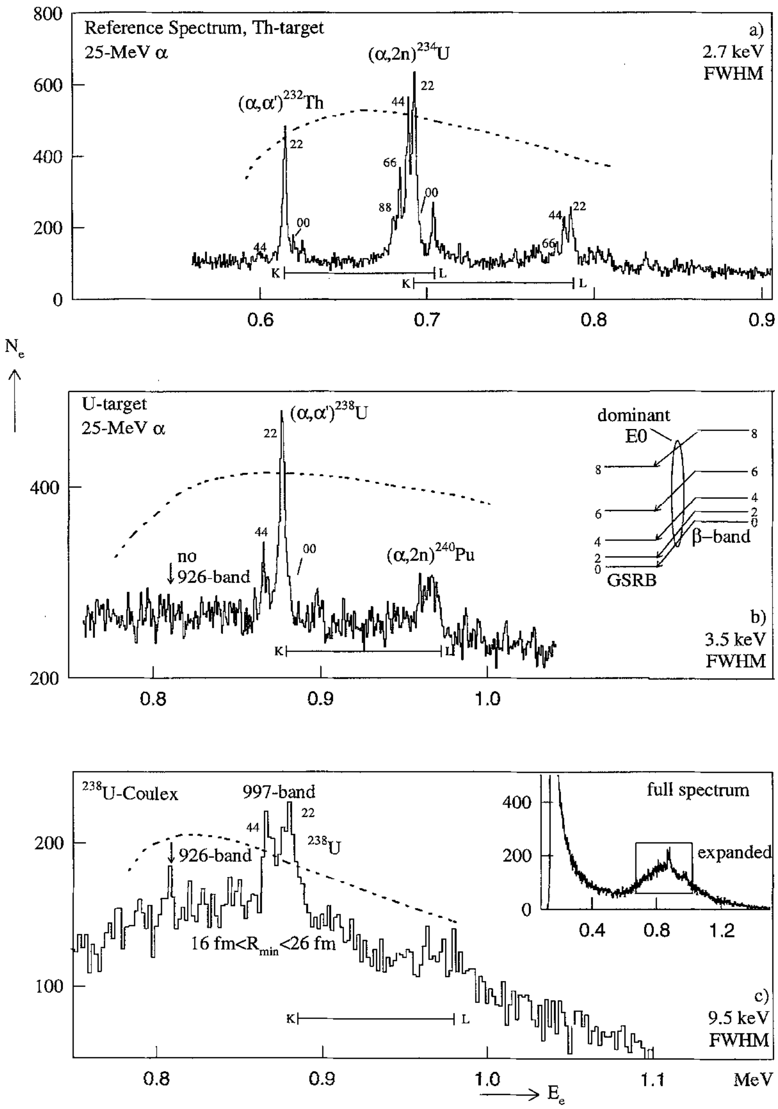

Fig. 1. Spectra of conversion electrons from E0-dominated $J \rightarrow J_g$ transitions as illustrated by the inset in figure b). The energy scale of reference spectrum a) taken with a Th target is shifted for easy comparison of the (α,α')- and (α,2n)-induced transitions in the following spectrum taken with a U target. Figure b) shows the spectrum with the best achieved energy resolution with a 0.5 mg/cm^2. U target. Spectrum c) is recorded with Coulomb excitation of ^{238}U projectiles and shows an enhanced 4 $\rightarrow$ 4_g intensity as compared to spectrum b) as well as a trace of the 2 $\rightarrow$ 2_g transition from the lower $K = 0$ (926 keV) band. However, spectrum c) shows no clear sign from higher-spin $J \rightarrow J_g$ transitions. The inset shows the full spectrum illustrating the effect of the Mini Orange transmission. The distance of closest approach R_{min} follows from the scattering angles of the U projectiles and the Ta ejectiles.

precise energy value E(0_3^+;^{240}Pu) = 1089.44(10) keV follows from the known $0_3 \rightarrow 2_g$ γ-ray transition [5].

Prior to our work the $0_3 \rightarrow 0_g$ E0 electrons of ^{238}U remained unobserved. The earlier studies yielded a rough value E(0_3^+;^{238}U) $\approx$ 992 keV from extrapolated energies of the 2^+ and 4^+ band members as deduced from γ-ray spectra and from particle spectra with poorer energy resolution. Our electron spectra yield an improved value E(0_3^+;^{238}U) = 996.7(6) keV. A fit to the narrow ^{240}Pu cluster locates the 2 $\rightarrow$

2_g transition at E = 967.5(8) keV, that is within error margins at its expected position slightly below the $0_3 \to 0_g$ transition. Spectrum 1c) has been recorded with Coulomb excitation of ^{238}U projectiles. After the corrections for Doppler broadening the ^{238}U cluster became visible on top of background of mainly δ rays with the already mentioned energy resolution of 9.5 keV, sufficient to separate the $2 \to 2_g$ and $4 \to 4_g$ transitions.

Typically observed halflives of the GSRB states range from 200 ps at $J_g^+ = 2^+$ to 20 ps at $J_g^+ \approx 10^+$. The partial halflives for in-band E2 transitions of states belonging to $K = 0$ bands will be similar, but their interband decay by $J \to J_g$ and $J \to J_g \pm 2$ transitions is much faster (≤ 1 ps) so that their intensities reflect the direct population of the 0_3-band levels. The relative K_{JJ}-line intensities give approximately the direct population of the states, since a J dependence of E0 admixtures will be minor [6] (less than 10% for $J \leq 6$, but increasing for higher spin values). Contrary to our expectation the higher-J transitions do not show up in the ^{238}U-Coulex and in the $(\alpha,2n)^{240}$Pu spectra. This is remarkable since Coulex measurements with Zr beams on ^{232}Th targets [7] yielded γ-ray spectra reaching β-band spins up to $J = 20$ with constant $J \to J_g$ intensities for spins up to $J = 8$. In the present case of ^{238}U the intensity $I(K_{44})$ increased relative to $I(K_{22})$ as compared to spectrum 1b), but the $6 \to 6_g$ remains invisible. Likewise no states with $J > 6$ are emerging in the ^{240}Pu spectra. As yet we do not see a full explanation for the absence of higher-spin states since it seems unconceivable that in these cases the collectivity gets lost with increasing spin. Perhaps the K line intensities of transitions from higher-spin states remain hidden in the background when the population of the states decreases rapidly with J, as Schuhbeck [8] found with Coulex by medium-heavy sulphur projectiles.

The spectra show levels of the *second* excited 0_3^+ bands, but not of the *first* $0_2^+(861;^{240}\text{Pu})$ band and only a 20% trace from the $0_2^+(926;^{238}\text{U})$ band in the case of the Coulex spectrum 1c). By comparing the noise level of the spectra with the intensity of transitions from 0_3 levels it is deduced that the $0_2^+(861;^{240}\text{Pu})$ band is absent to within at least a factor of two (quoted in Table 1, but just outside Fig. 1b) with respect to the observed 0_3 band members. However, the existence of the *first* excited 0_2^+ states is

Table 1
Energy and intensity values.

	$J_i^+ \to J_g^+$	$E(J_i^+)$ (keV)	Intensity K-line
^{238}U-0_2^+		925.7(3)*	Trace after Coulex; $I_{22}(0_2^+) \approx I_{22}(0_3^+)/5$
^{238}U-0_3^+	$0^+ \to 0^+$	996.7(6)	0.22(5) after (α,α')
	$2^+ \to 2^+$	1037.4(2)*	≡1 (norm)
	$4^+ \to 4^+$	1130.6(4)	0.3(1) after (α,α'); 0.5(1) after Coulex
^{240}Pu-0_2^+		860.71(10)*	not observed; $I_{22}(0_2^+) < I_{22}(0_3^+)/2$
^{240}Pu-0_3^+	$0^+ \to 0^+$	1089.44(10)*	not observed, but assumed from known $\gamma(0_3^+ \to 2_g^+)$ [5]
	$2^+ \to 2^+$	1130.94(10)	≡1 (norm)
	$4^+ \to 4^+$	1226.6(13)	0.8(4)
	$6^+ \to 6^+$	1375.5 (7)	0.8(4)

* from [5]

known [5] from particle and γ-ray spectra recorded after reactions with charged particles and neutrons, and the 0_2^+ band levels will have been populated as well during the present experiments. We explain their absence (^{240}Pu) or near invisibility (^{238}U) in the electron spectra as due to a smaller E0 admixture and less enhanced K conversion. The E0 admixture can be estimated in the Rotational Vibrational Model [9] (RVM) parameterization from the matrix elements $\langle 0_\beta^+ || M(\text{E0}) || 0_g^+ \rangle = A^2 \beta_0^2 \sqrt{2E(2_g^+)/E(0_\beta^+)}$ and $\langle 0_{K=0}^+ || M(\text{E0}) || 0_g^+ \rangle = A^2 \beta_0^2 E(2_g^+)/E(0_\gamma^+)$, where $A = (3Ze/4\pi) R_0^2$ and where β_0 and R_0 are associated with the equilibrium deformation and the radius of the nucleus, respectively. The first matrix element referring to the 0_3^+ band and the second one to the 0_2^+ band. The E0 enhancement appears to be less by a factor of six (^{238}U) or five (^{240}Pu) for the 0_2^+ bands.

Fig. 2 shows the moments of inertia of the GSRB and 0_2^+ bands as deduced from the Nuclear Data Sheets [5] and of the 0_3^+ bands determined by the present work. In the case of ^{238}U the $0_2^+(926)$ curve starts with a remarkably steep gradient assuming that the level energies and assignments of this 0_2 band are accepted as correct. From the incorrect old bandhead energy [5] of 992 keV would follow a similarly extreme steepness for the ^{238}U 0_3^+ curve. The pattern of moments of inertia $\mathcal{J}(I_n^+)$ depends critically on the values of $E(0_n^+)$ and $E(2_n^+)$. Our improved value $E(0_3^+)$

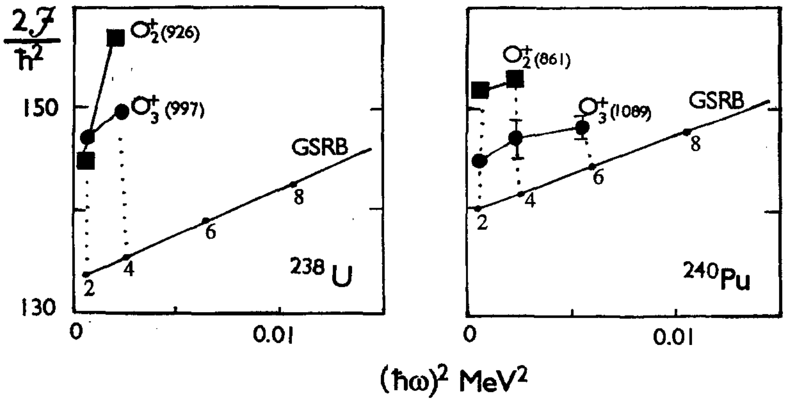

Fig. 2. Increase of moments of inertia $\mathcal{J}$ with rotational frequency ω for $K = 0$ bands in ^{238}U and ^{240}Pu. Uncertainties do not exceed the size of the data symbols except for the 0_3^+ band of ^{240}Pu.

$= 996.7(6)$ keV yields, together with a value $E(2_3^+) = 1037.4(2)$ keV known from γ-ray spectra, the beginning of a curve with a slope similar to that of the GSRB.

The $(\alpha,2n)^{240}$Pu fingerprint of a narrow cluster appears in analogy with the reference $(\alpha,2n)^{234}$U spectrum and indicates that the GSRB- and 0_3^+-band members of ^{240}Pu have, likewise those of ^{234}U, not very different moments of inertia. They approach the phenomenon of identical bands; a conclusion which is allowed despite poor statistics and a by consequence somewhat tentative cluster analysis. With the linewidth of ≈ 3.5 keV the cluster decomposes in three ^{240}Pu K lines ($2 \to 2_g$, $4 \to 4_g$ and $6 \to 6_g$; Table 1) just underneath the $L(1037;^{238}\mathrm{U})$ transition. Though the spectra are "noisy", the existence of the ^{240}Pu cluster is undisputed; it has been reproduced as an unresolved but clearly broadened peak in other spectra with higher beam current and consequently poorer resolution. The intensities quoted in Table 1 are estimated from the combined spectra.

The appearance of two excited 0^+ states in close proximity at lower energies in ^{238}U and ^{240}Pu is difficult to accommodate in theoretical models. Searching for additional clues we inspected the γ-ray branching ratios of stretched E2 transitions from 2^+ states of $K = 0$ bands:

$$R = \frac{B(\mathrm{E2}; 2^+_{\beta\,\mathrm{or}\,\gamma} \to 4_g^+)}{B(\mathrm{E2}; 2^+_{\beta\,\mathrm{or}\,\gamma} \to 0_g^+)}. \quad (1)$$

This ratio can be deduced for a number of accessible

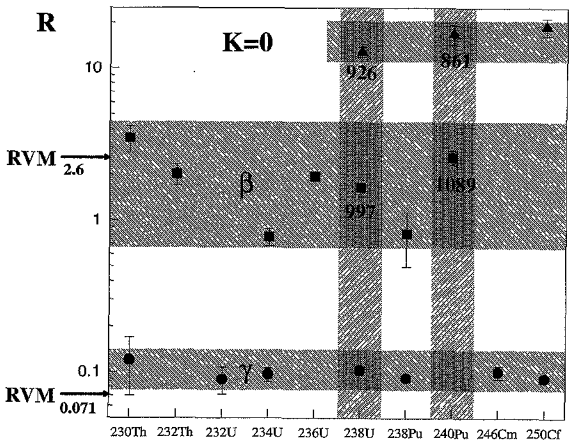

Fig. 3. Overview of γ-ray branching ratios R from 2^+ states of $K = 0$ bands and of γ-bands with $R = B(\mathrm{E2};2 \to 4_g)/B(\mathrm{E2};2 \to 0_g)$. Left-side arrows indicate RVM-values without mixing. $R(0_3^+)$ values of ^{238}U and ^{240}Pu belong to the regular middle zone, while their $R(0_2^+)$ values and a literature value of ^{250}Cf [10] are much larger. When the error bars are not shown they are smaller than the marker size.

actinide nuclei [5] and does not concern the $2^+_{\beta\,\mathrm{or}\,\gamma} \to 2_g^+$ transitions which would introduce complicating E2-M1-E0 mixing. The overview of Fig. 3 shows as a surprise that the ratio R is very different for the 0_3^+ and 0_2^+ bands. For comparison the figure includes γ-band values. They fall in a narrow zone with RVM quantum numbers [9] $K = 2$, $n_0 = n_2 = 0$ somewhat above the RVM value $R_\gamma = 0.071$ without mixing. The regular $K = 0$ values with $n_0 = 1$, $n_2 = 0$ show more spreading but can be grouped in a zone which encompasses the RVM value $R_\beta = 2.6$. The $R(0_3^+)$ values for ^{238}U and ^{240}Pu, respectively 1.66(6) and 2.6(3), lie in this zone. However, the $R(0_2^+)$ values are much larger: 13(1) and 17(3), respectively. Together with a third value of 19(3), which we traced for ^{250}Cf [10], they may form a third zone. It is tempting to relate this zone to $K = 0$, $n_0 = 0$, $n_2 = 1$, although we believe that R depends to first order only on J and K. Recent calculations using the Geometrical Collective Model (GCM) [11] are able to yield a variety of R values close to the respective β- and γ-band values. Furthermore, this GCM gives some evidence for much larger R values of 2^+ states. However, the exact structure of these states is not yet clear.

A consistent theoretical description of the actinides has been pursued by various investigators with varying degrees of success. The observed $K = 0$ states were first interpreted as collective quadrupole vibrations; mostly β vibrations with preserved axial symmetry, but possibly also as triaxial γ vibrations with two phonons combining to a 0^+ state. Davydov and Rostovsky [12] already pointed out that E0 admixture to $2^+ \to 2_g^+$ transitions will be more pronounced for β bands with rigid axial symmetry than for triaxial γ bands. For a wide range of nuclei they find:

$$\frac{B(\mathrm{E0}; 2_\beta \to 2_{\mathrm{gs}})}{B(\mathrm{E2}; 2_\beta \to 2_{\mathrm{gs}})} \gg \frac{B(\mathrm{E0}; 2_\gamma \to 2_{\mathrm{gs}})}{B(\mathrm{E2}; 2_\gamma \to 2_{\mathrm{gs}})}.$$

In our cases the E0 enhancement of all $J \to J_g$ transitions with $J > 0$ will decrease with increasing triaxiality. In this perspective the 0_2^+ states, which were hardly observed in the electron spectra, are triaxial while the 0_3^+ levels are axially symmetric. However, the remaining problem is that triaxial γ bands are expected to manifest themselves at higher energies. That deformed heavy nuclei exhibit multiple $K = 0$ bands below the pairing gap remains an item of controversy in the theoretical interpretations, with different approximations leading to opposite conclusions. On one side Soloviev et al. [13] expressed as their opinion that collective two-phonon excitations (e.g. double-γ or double-β vibrations) *will not exist* in actinides below 2 MeV. They infer that this is due to Pauli blocking of important quasi-particle components. Instead, they describe the first 0^+-excitations in ^{238}U and ^{240}Pu with the quasi particle-phonon nuclear model (QPNM). By using not only particle-hole but also particle-particle interactions they were able to produce for these nuclei two low-lying 0^+ states between the 0.8 and 1.2 MeV. On the other side, Piepenbring and Jammari [14] arrive at the rather opposite conclusion. By using a "multiphonon method" they find that two-phonon excitations *retain* a collective character. Experimental evidence for the existence of two-phonon states has been given by Börner et al. [15] in the rare-earth region for ^{168}Er.

Strong pair correlations in excited 0^+ states of actinide nuclei are known from (p,t) transfer reactions [16]. Jänecke et al. [17] observed in the actinides a strong α-cluster pickup with (d,^{6}Li) reactions suggesting coherent contributions from both neutron and proton pair excitations. They present an overview of experimental 0^+ excitation energies as a function of the total number of bosons (IBM model) outside ^{208}Pb. It is interesting that in this overview the excited 0_3^+ states of ^{238}U and ^{240}Pu fall in one group while the two second excited 0_2^+ states fall in another group.

Understanding the structure of the heaviest, densest and still accessible nuclear systems will further improve with future experimental and systematic efforts to accumulate spectra with good energy resolution and improved statistical accuracy. However, with the achieved spectroscopy of conversion electrons, the ^{238}U and ^{240}Pu findings already lead to the following conclusions:

- The spectra demonstrate conclusively the existence of bands based on two excited 0^+ states which are nearby in energy, but widely different in structure.
- The spectra show E0-enhanced $J \to J_g$ transitions from the 0_3^+, but less or not at all from the 0_2^+ bands. These 0_2^+ bands most likely exhibit less E0 admixture and are as a result less converted. It is tempting to treat them in terms of triaxiality.
- The γ-ray branching ratio R is larger by one order of magnitude for the anomalous 0_2^+ band as compared with the 0_3^+ one.
- The observed 0_3^+ band for ^{240}Pu shows moments of inertia which are approaching those of the GSRB.

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Faessler for correspondence about E0 and E2 matrix elements.

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