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EXPERIMENTAL INFORMATION CONCERNING DEFORMATION OF NEUTRON RICH NUCLEI IN THE A∿100 REGION*

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Experimental results are presented on the ground state bands of light even-even nuclei produced in the primary fission of ^{252}Cf. The systematics of the energy spacings and life times are similar to those of deformed nuclei in the rare earth and actinide regions.

In this letter we report experimental evidence for rotational-like behavior in very neutron rich even-even $_{40}$Zr, $_{42}$Mo, $_{44}$Ru, and $_{46}$Pd isotopes. These results support recent theoretical studies by Ragnarsson and Nilsson[1] and by Arseniev _et_ _al_.[2] which have predicted a new region of stable deformation which includes these nuclei. Fission fragments from spontaneous fission of ^{252}Cf provided experimental access to this region. We have obtained systematic information on the ground state bands of all the light even-even fission products having calculated independent yields[3] of greater than approximately 1% per fission.

Prompt K x-rays and/or γ-rays in coincidence with pairs of fission fragments were measured using the detector arrangement indicated in Fig. 1. Three separate experiments using different photon detectors were performed: 1) recording γ-rays with a 1 cm^3 Ge(Li) detector (resolution 1 keV at 122 keV) in position γ2; 2) recording γ-rays and/or x-rays in coincidence using a 6 cm^3 Ge(Li) detector in position γ1 and a 2 cm^2 Si(Li) detector in position

$\gamma 2$; 3) recording γ-ray, γ-ray coincidences with a 35 cm^3 Ge(Li) coaxial detector in position $\gamma 2$ and a 6 cm^3 Ge(Li) detector in position $\gamma 1$. In all the experiments a nominally 10^5 fission per minute source of ^{252}Cf was electrodeposited onto the surface of fragment detector F1. Thus Doppler shifting and broadening problems were eliminated for transitions from the fragments stopped in that detector. This technique, which simplified the spectra applies to half life times longer than the stopping time of the fragments ($\sim 10^{-12}$ sec). Life time determinations in the time region 0.1 - 2.0 nsec were obtained from the ratio of the non-Doppler shifted gamma ray intensity observed when the fragment stopped in the plated detector F1 relative to the intensity observed when the fragment stopped in the second detector F2, which was separated from the plated detector by 8 mm. The various detector systems were digitally gain stabilized using external gamma ray sources as indicated in Fig. 1.

In all the experiments the analog pulse heights were digitized and stored event by event in a PDP-9 computer. The on-line computer was programmed to monitor the resolution of the detectors and to transfer the experimental data onto magnetic tape in a compressed format. A total of 2×10^8 multiparameter events were recorded and later processed on a CDC 6600 computer.

The masses of the fragments were calculated from the measured energies using the Schmitt calibration method and the known neutron corrections.[4] Gamma-ray spectra associated with fragment masses in 2 amu wide mass intervals were obtained by sorting the three parameter data. Each of these spectra was then analyzed to give quantitative energies and intensities of individual transitions. This was accomplished using the on-line photopeak analysis code developed by Routti and Prussin.[5] The widths of the mass distributions associated with single gamma transitions ranged from 4.0 to 6.5 amu (FWHM) and the mean values of the masses for these distributions were determined with standard statistical errors of less than 0.2 amu for the strong transitions; however, the absolute determination of the masses are uncertain by ± 1 amu due to systematic errors in the calibration procedure and/or the neutron corrections. The x-ray, gamma-ray coincidence data were used to obtain definite Z assignments for the observed transitions. The gamma, gamma coincidence data were then used to obtain information on additional transitions associated with single isotopes.

Since it was to be expected that the radiations associated with even-even isotopes should come from low lying states following a simple systematic behavior, our study began with the investigation of these nuclei. From analysis of the data we have been able to assign transitions to 12 even-even isotopes for which no previous assignments have existed. The results of the investigation are summarized in Table I. For each isotope in the table we present two lines of information. The top line contains the experimental energies of the observed levels along with the ratio of the energies of the $4^+/2^+$, the measured half life of the 2^+ level, and the yield per fission of this transition. Also presented are the calculated $B(E2;2 \rightarrow 0)$ and β_2 values following the formalism of Stelson and Grodzins.[6] The second line contains corresponding predicted values. Several criteria were taken into account in making these assignments.

1. The intensities of the $2 \rightarrow 0$ ground state band transitions corrected for internal conversion follow the calculated independent fission yields of the even-even isotopes.[3] Such a correspondence would be expected on the basis of considerations involving the removal of the initial 6 - 10 units of angular momentum associated with each fragment. The decay sequence

should be analogous to the prompt decay of even-even products in (particle, xn) reactions in which the ground state band is fed very strongly. The calculated independent yields are based on empirical information concerning the most probable charges (Z_p) of the mass chains and the charge distribution about this value. They are not believed to be absolutely accurate but should be reasonably good estimates.

2. The masses associated with the 2 → 0 transitions of the even-even isotopes as determined from the kinetic energies of the fragments are within ± 1 amu of the assigned masses. We were also able to obtain mass assignments for transitions from odd Ru nuclei and one Zr isotope which are between the masses of the isotopes assigned as even-even.
3. The energies of the 2^+ levels obey smooth systematics. The trends show a decrease in the 2^+ level energies with increasing displacement from the closed shells Z = 50 N = 50. An exception is the increase in the energy of the 2^+ state of ^{116}Pd relative to ^{114}Pd which may be due to the influence of the N = 82 shell.
4. All even-even isotopes with prompt yields > 1.0% are seen. There are no missing cases.
5. The multipolarity of the transition assigned as $2^+ \rightarrow 0^+$ in ^{110}Ru was found to be E2 in studies of electron conversion.[4] From other previous studies which measured anisotropies of prompt gamma rays, multipolarities of the $2^+ \rightarrow 0^+$ transitions in ^{106}Mo and ^{110}Ru were found to be consistent with E2 transitions.[7]
6. High spin members of the ground state bands have been extracted from γ-γ coincidence data. The transitions assigned as 4 → 2 have the highest intensities in the spectra taken in coincidence with the 2 → 0 transitions. Knowledge of the 2^+ and 4^+ members of the band allow the other members to

be predicted with good accuracy. These predictions can be obtained empirically[8] as we have done or through analyses using one of the various two parameter moment of inertia models. The predicted results are shown below the experimental data in Table I. For transitions with sufficient intensity to permit observation this gives additional confidence in the assignment. The ratio of the energies of the $4^+/2^+$ is an indication of the softness of the nucleus and these values show smooth trends between adjacent even-even nuclei.

7. The exact Z assignments were made for most of the transitions from the present measurements of K x-rays in coincidence with gamma rays and by comparison with earlier work of Watson[4] on measurements of K x-rays in coincidence with conversion electrons. For a γ-ray, x-ray coincidence it is necessary to have a γ-ray cascade with at least one of the members undergoing internal conversion in the K electron shell. This implies that the $2 \rightarrow 0$ ground state γ-ray transition will not be strongly observed in coincidence with x-rays since it requires the internal conversion of a higher energy member of the cascade; however, the $4 \rightarrow 2$ transitions can be seen since their observation depends on the probability for internal conversion of the lower energy $2 \rightarrow 0$ transition. We have seen the $4 \rightarrow 2$ transitions in coincidence with K x-rays for the isotopes ^{100}Zr, ^{102}Zr, ^{104}Mo, ^{106}Mo, ^{108}Ru, ^{110}Ru. We also saw $2 \rightarrow 0$ transitions for some of the isotopes ($^{(96)}$Sr, ^{100}Zr, ^{102}Zr, ^{104}Mo, and ^{106}Mo) by observing the transitions in coincidence with the K x-rays of the complementary fragment. The complementary fragment of an even-even product can have odd neutron numbers (due to a distribution in the number of neutrons evaporated) and can therefore have a high probability of emitting a K x-ray following internal conversion of one of its low energy transitions.

8. For all of the isotopes in Table I, with the exception of ^{112}Ru, the $2 \rightarrow 0$ ground state transitions were also observed in a previous experiment which followed beta decay of the unseparated prompt products.[9] These $2 \rightarrow 0$ transitions observed following beta decay were seen with appropriate half lives and with intensities which are a substantial fraction (>50%) of the calculated cumulative mass chain yields.
9. Our measurements of the life times are believed to be uncertain by as much as 20% and in principle our values represent upper limits since there is the possibility of hold-up in the previous transitions. However, comparison of these results with values predicted using the empirical relationship of Mariscotti *et al.*[10] shows that the agreement is perhaps better than could be expected.

Single particle (B(E2;$2 \rightarrow 0$) values and the theoretical deformation values of Arseniev *et al.*[2] are presented in Table I below the corresponding quantities derived from the experimental data. The theoretical calculations predict this to be a region for which the equilibrium shape is an axially symmetric oblate spheroid. We have translated the reported ε_0 deformation to β_2 deformations ($\beta_2 \cong \varepsilon_0/0.95$).

The central question from these studies is whether the theoretical predictions for deformation can be verified. It is not possible to determine the existence of static deformations from observed energy level spacings or from measurements of B(E2;$2 \rightarrow 0$). However, studies of such systematics are indicative of nuclear softness and therefore it is of interest to compare these properties in this new region with the corresponding values for the rare earth and actinide regions which are the two major areas of known permanent deformation. There are several different indicators of deformation and it is informative to compare each. Fig. 2 is a composite plot containing five indicators associated with deformation [β_2, $B(E2)/B(E2)_{SP}$, E_{4^+}/E_{2^+}, β_2/β_{2SP}, $(79.51/E_{2^+}) \times (158/A)^{5/3}$] plotted as a function of mass. The last indicator represents to a first approximation a mass independent comparison of the energies of the first 2^+ states using arbitrarily the deformed ^{158}Gd nucleus as a reference. The nuclei presented in the plot include the current region (96 - 116), and a representative sampling of isotopes in the rare earths (150 - 180) and in the actinides (224 - 244).

In this light fission-product region, of the isotopes studied, ^{102}Zr appears as the most favorable candidate for deformation. Its values for β_2 (0.604) and for the mass independent energy parameter (1.08) are larger than any of the corresponding values found in the rare earth and actinide nuclei. Also its values for $B(E2)/B(E2)_{SP}$ (234.) and β_2/β_{2SP} (15.2) are larger than for any of the rare earths though smaller than some of the actinides. The only parameter for which it has a lower value than obtained in the other regions is the E_{4^+}/E_{2^+} ratio where the ^{102}Zr value of 3.15 is somewhat smaller than the limiting value for a perfect rotor (3.33) which is closely approached in both the rare earth and actinides. The other new isotopes for which we present information have smaller values for these deformation indicators than ^{102}Zr but even they have, in several instances, values comparable or larger than those typically found in the rare earth and actinide region and in all cases are larger than the values found for spherical nuclei near closed shells.

For the isotopes with higher masses the decrease in the deformation indicators is believed to be due to the approach of the Z = 50 closed shell, and for the lighter isotopes the effect of the N = 50 shell should be important. The theoretical calculations of Arseniev *et al*. imply that the regions of strongest deformation should be in the heavier isotopes of strontium (98 -

102) and of krypton (96 - 102) which are not produced in significant yield in the fission process. Recently (t,p) reactions leading to ^{98}Zr have shown its lowest 2^+ state to be at 1.223 MeV.[11] Unless ^{98}Zr has a lower lying collective state which has not been detected, the change in the energy of the lowest 2^+ states between ^{98}Zr and ^{100}Zr is larger than the equivalent change from ^{150}Sm to ^{152}Sm.

We are grateful to the following persons for their help in this work: Elizabeth Quigg wrote the necessary programs for the PDP-9 computer. Thomas Strong handled the processing of our data using the CDC 6600 computer. Robert Latimer and James Harris electrodeposited the ^{252}Cf sources on our fission detectors. Very useful discussions with John Rasmussen, Chin Fu Tsang, Frank Stephens, and Rand Watson are acknowledged.

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FOOTNOTES AND REFERENCES

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FIGURE CAPTIONS

Fig. 1 Schematic representation of detector system. Detectors F1 (with electrodeposited ^{252}Cf) and F2 measured energies of fragments. Detectors γ_1 and γ_2 measured energies of γ-rays and/or x-rays. External sources for stabilization of the photon detectors were ^{243}Cm (α-γ coincidence), ^{60}Co (γ-γ coincidence) and ^{241}Am (α-γ coincidence).

Fig. 2 A composite plot containing five indicators of deformation plotted as a function of mass. The mass intervals used contain only the current experimental region (96 - 116) and a representative sampling from the two major known regions of deformation. The rare earth and actinide data were taken from refs. 5 and 9. The values of β_2, β_{2SP}, B(E2), and B(E2)$_{SP}$ were extracted from relationships presented in ref. 6; E_{2^+} and E_{4^+} are the experimental energies of the first 2^+ and 4^+ levels; the final indicator $(79.51/E_{2^+}) \times (158/A)^{5/3}$, gives a relative comparison between the energies of the first 2^+ states on a basis which removes the inherent mass-dependence from the moment of inertia. The open circles represent current results obtained using experimental energies and life times. The open squares represent current results obtained using experimental energies and calculated life times (ref 10). The closed circles represent literature values (refs 6 and 10).

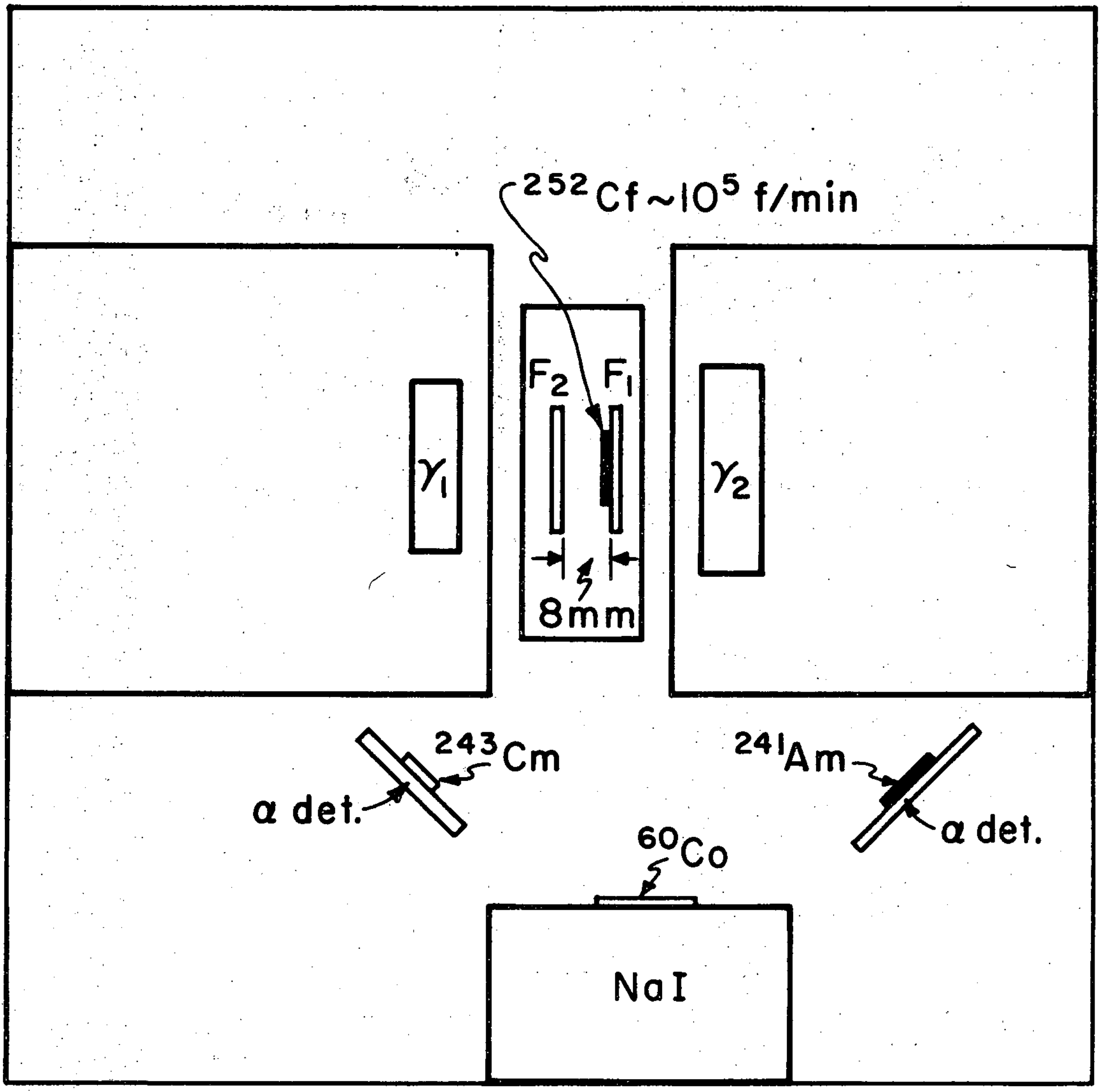

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Fig. 1

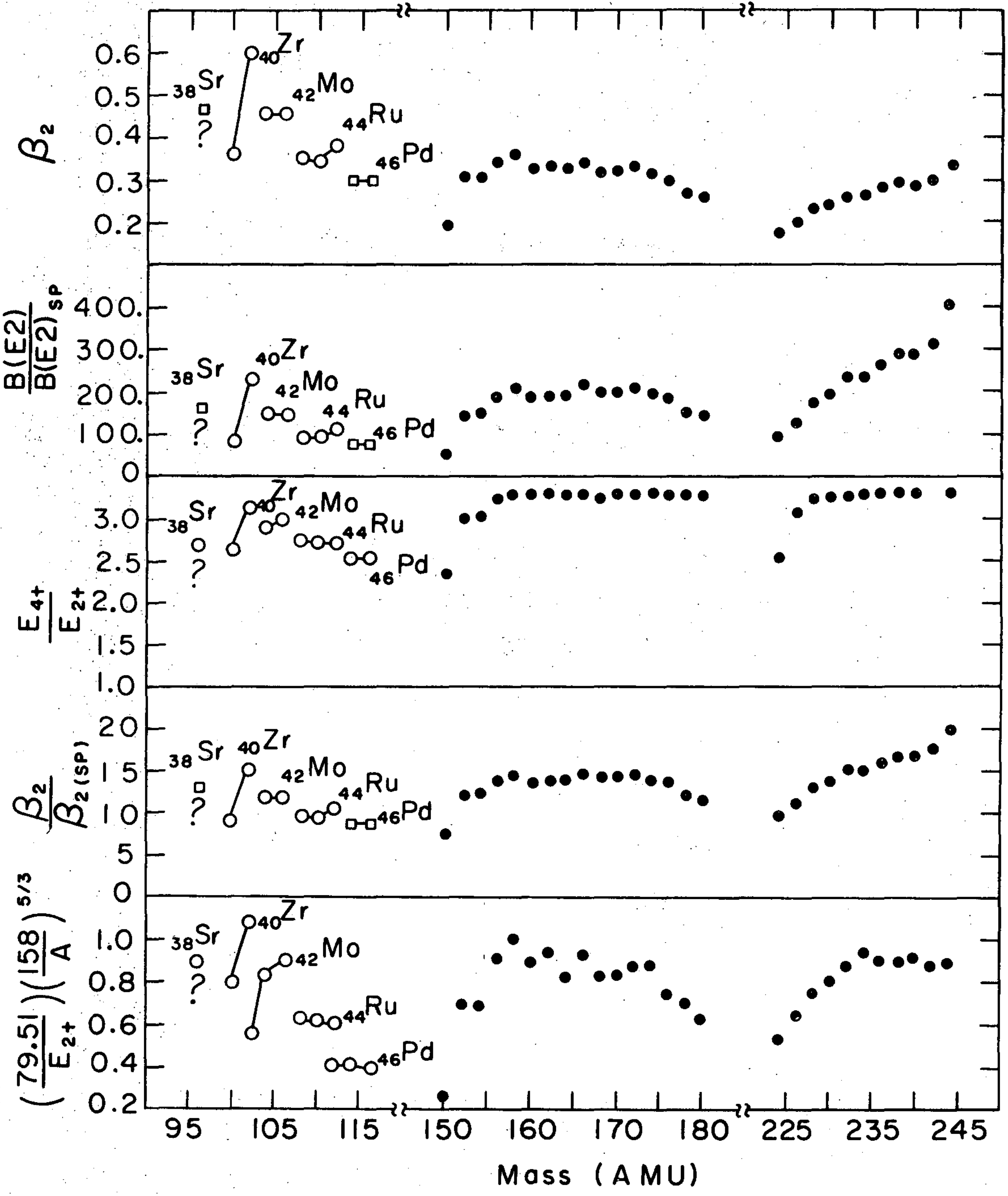

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Fig. 2

Table I. Experimental results and phenomenological predictions for ground state bands.

		Energy in keV				E4/E2	$t_{1/2}(2\rightarrow 0)$	Yield[a]	Mass	B(E2; 2 → 0)[b]		β_2[c]
		2^+	4^+	6^+	8^+		nsec	%/fis		exp	s.p.	
$^{(96)}$Sr[d]	exp	(204.1)	(556.3)			2.72	<1.7	0.51	96.0	>87		>0.24
	pred			1029	1582		0.34	0.67	(96)		2.59	
^{100}Zr	exp	212.7	564.8	1062.7		2.65	0.52	1.80	100.53	233		0.364
	pred			1021	1563		0.29	1.82	100		2.74	-0.29
^{102}Zr	exp	151.9	478.5	964.5	(1551)	3.15	0.86	1.43	101.85	658		0.604
	pred			949	1533		0.94	0.82	102		2.81	-0.29
^{102}Mo	exp	296.0					<0.1	0.46	103.04	>241		>0.348
	pred							0.82	102		2.81	
^{104}Mo	exp	192.3	561.0	1081.0		2.92	0.45	3.37	104.67	430		0.459
	pred			1075	1681		0.45	3.12	104		2.88	-0.28
^{106}Mo	exp	171.7	522.5	(1034.3)		3.04	0.75	3.37	106.04	433		0.454
	pred			1008	1604		0.61	2.49	106		2.96	-0.27
^{108}Ru	exp	242.3	665.3			2.75	0.22	1.94	108.99	293		0.353
	pred			1240	1914		0.17	2.73	108		3.03	-0.26
^{110}Ru	exp	240.8	663.9	1240.0	(1947.7)	2.76	0.23	3.49	110.15	289		0.346
	pred			1238	1914		0.18	3.25	110		3.11	-0.25
^{112}Ru	exp	236.8	645.7			2.73	0.20	0.97	111.85	361		0.382
	pred			1200	1847		0.19	0.70	112		3.18	-0.25
^{112}Pd	exp	348.8					<0.1	0.77	112.90	>108		>0.199
	pred							0.71	112		3.18	
^{114}Pd	exp	332.9	853.6	1503.0		2.56	<0.1	1.48	114.36	>136		>0.221
	pred			1515	2304		0.052	1.77	114		3.26	
^{116}Pd	exp	340.6	878.6			2.58	<0.1	0.87	115.25	>121		>0.207
	pred			1570	2384		0.045	0.73	116		3.34	

[a] Yield; exp. - no. of 2 → 0 transitions per fission corrected for internal conversion; pred. - radio-chemical yield of g.s. (see text).

[b] B(E2) are in units of $e^2\ cm^4 \times 10^{-51}$.

[c] The experimental β_2 values are derived from the B(E2) data following Ref. 6. The sign is undetermined.

[d] The assignments of the levels as 2^+ and 4^+ associated with A = 96 are uncertain. The transition assigned as 4 → 2 has a life time of >1 nsec but may be partially held up by a previous delayed transition.

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