

ANL/PHY/CP-89274

NUCLEAR STRUCTURE AND SHAPES FROM PROMPT GAMMA RAY  
SPECTROSCOPY OF FISSION PRODUCTS

CONF-961010--2

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SEP 03 1996

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**Abstract:** Many nuclear shape phenomena are predicted to occur in neutron-rich nuclei. The best source for the production of these nuclides is the spontaneous fission which produces practically hundreds of nuclides with yields of greater than 0.1% per decay. Measurements of coincident gamma rays with large Ge arrays have recently been made to obtain information on nuclear structures and shapes of these neutron-rich nuclei. Among the important results that have been obtained from such measurements are octupole correlations in Ba isotopes, triaxial shapes in Ru nuclei, two-phonon vibrations in  $^{106}\text{Mo}$  and level lifetimes and quadrupole moments in Nd isotopes and  $A=100$  nuclei. These data have been used to test theoretical models.

**1. Introduction**

Nuclear structure and nuclear shape are governed by the residual interactions whose magnitude depends on the single-particle states near the Fermi level. Thus the shape and structure of nuclei are functions of proton number and neutron number. Most nuclear reactions produce nuclides which are more neutron deficient than the nuclei along the line of  $\beta$  stability and hence a large amount of knowledge is available on structures and shapes of stable and neutron deficient nuclei. Until recently, data on neutron-rich nuclei were sparse, mainly because the mechanisms which produce neutron-rich nuclei produce a large variety of nuclei simultaneously. Individual nuclei could not be studied in the presence of gamma rays generated by other abundant nuclides. The most efficient source for the production of neutron-rich nuclei is fission. Since hundreds of nuclides are produced in fission, it is impossible to study transitions in one nucleus in the presence of  $\gamma$  rays from other products. Studies of such nuclides can only be made if some device is used to isolate individual nuclides or sensitive instruments are developed to observe transitions in one particular nucleus. The structure information can be obtained either from the measurements of radiations from the  $\beta^-$  decay of the secondary fragments and its daughters or by measuring the prompt  $\gamma$  rays deexciting the secondary fragments<sup>1</sup>. Since the half lives of fission products range from seconds to years,  $\beta^-$  decay studies can be carried out by either chemically separating the nuclides of interest or by using isotope separators to produce samples. For the second type of measurements, prompt  $\gamma$  rays from fission source are measured. With the recent availability of large arrays<sup>2</sup> of Ge detectors, such measurements are possible and this article describes these measurements and results obtained from them. The techniques for the determination of nuclear structures of fission products with large Ge arrays and the results from these experiments have recently been reviewed by Ahmad and Phillips<sup>1</sup> and Hamilton et al<sup>3</sup>.

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The first experiment to study the structure of fission products was performed by the Berkeley group<sup>4,5</sup> who used one Ge(Li) detector to measure  $\gamma$  rays in coincidence with fission fragments. Two Si detectors were used to measure the kinetic energies of the coincident fragments, from which the fragment masses were deduced. In this way, the  $\gamma$  rays were assigned to individual nuclides and the first few levels of the ground bands were determined. This experiment provided systematic information on the ground-state bands of many even-even nuclei in the mass 100 and mass 140 regions with independent yields of greater than 1%. The data showed for the first time that many of the very neutron-rich even-even isotopes of Zr, Mo, Ru and Pd were deformed. The field was dormant until the mid-eighties when large arrays of Compton-suppressed large volume Ge detectors became available. The first experiment with a large Ge array was performed at Argonne<sup>6</sup> with a <sup>252</sup>Cf source which demonstrated that even two-fold coincidences can produce fairly clean spectra of individual nuclides. Using the earlier information obtained by the Berkeley group, Phillips et al.<sup>6,7</sup> were able to determine the structures of even-even Ba and Ce isotopes up to spin  $15 \hbar$ . The data showed that <sup>144</sup>Ba, <sup>146</sup>Ba and <sup>146</sup>Ce were octupole deformed, in agreement with the predictions of theoretical models<sup>8,9</sup>. Since then many experiments have been performed at several laboratories with different fission sources. We have used a <sup>248</sup>Cm source and performed experiments with the Argonne Notre Dame BGO array and the EUROGAM array. In the following we discuss the latest experiment with the EUROGAM II array<sup>10</sup> and present some results obtained from the analysis of this data set.

The aim of the studies of the fission products is to obtain structures of very neutron-rich nuclei so that one may determine the influence of isospin on nuclear structure. The  $N/Z$  ratios of the fission fragments are practically the same as that of the fissioning nucleus. Thus to access most neutron-rich nuclides, fission sources with the largest  $N/Z$  ratios should be used. Spontaneous fission produces more neutron-rich fragments than fission induced by nuclear reactions because in nuclear reactions more neutrons are evaporated before the formation of the final fragments. However, nuclear reactions can be used to enhance the yields of certain nuclides because their mass distributions are different from those of nuclides which decay by spontaneous fission. The  $N/Z$  ratios of the most favorable fissioning systems and the most probable mass of the primary fragment<sup>11</sup> are presented in table 1.

Table 1. Most probable mass of primary Zr fragments

Fissioning System	$N/Z$ Ratio	Primary Zr Mass <sup>a</sup>
<sup>233</sup> U( <i>n</i> <sub>th</sub> , <i>f</i> )	1.543	100.5
<sup>235</sup> U( <i>n</i> <sub>th</sub> , <i>f</i> )	1.565	101.3
<sup>238</sup> U( <i>n</i> , <i>f</i> )	1.598	102.6
<sup>248</sup> Cm( <i>sf</i> )	1.583	102.0
<sup>252</sup> Cf( <i>sf</i> )	1.571	101.6
<sup>254</sup> Cf( <i>sf</i> )	1.592	102.4

<sup>a</sup>Most probable primary mass is given by  $A = (A_f/Z_f)(Z-0.5)$  (Unik et al.).

Mass distributions in the fission of these nuclides have been measured and for <sup>248</sup>Cm, the source used in our experiments, the mass distribution<sup>12</sup> is shown in Fig. 1. As can be seen, the yields are maximum for mass around 100 and mass around 144 with yields of ~6%. For each mass, the yield is distributed among several elements. Systematics<sup>13,14</sup> of charge distributions have been determined for many fissioning systems and from these data a universal curve has been derived (Fig. 2). By combining the two distributions, it is possible to determine the independent yields of individual nuclides.

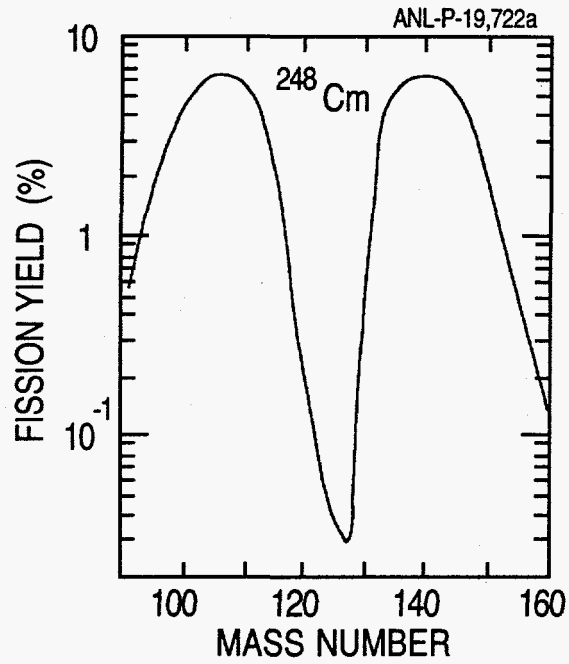


Fig. 1. Mass distribution in the spontaneous fission of  $^{248}\text{Cm}$  (Flynn et al.). The yields are normalized to 200%.

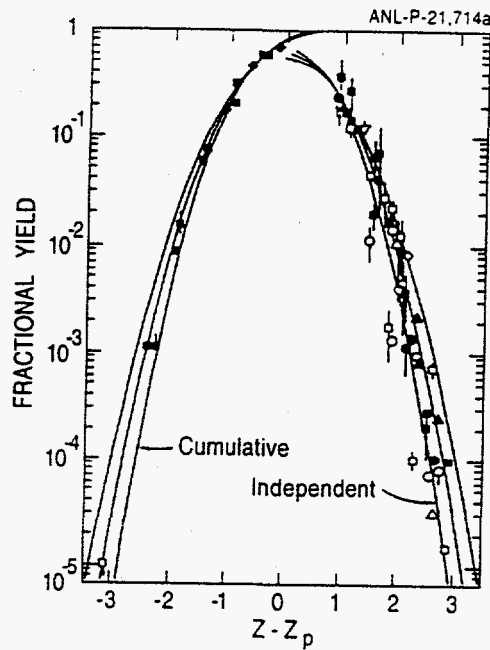


Fig. 2. Charge distributions for several fissioning systems (Wahl et al.). Fractional yields are plotted against  $Z - Z_p$ , where  $Z_p$  is the most probable charge and its value is derived from an empirical fit. The curves are calculated with  $\sigma_z = 0.62 \pm 0.06$ .

## 2. Experimental Measurements

### 2.1. Fission Source

Fission fragments have kinetic energies of  $\sim 1$  MeV/u and the range of these fragments in different materials is between 3 and 15 mg/cm<sup>2</sup>. The fission source thickness, if it were a pure material, is too small to stop the fragments in the source. Consequently the  $\gamma$  rays will be Doppler broadened. In order to avoid this broadening of  $\gamma$ -ray peaks, the fragments should be stopped within the source. Since we are also interested in observing K- X-rays from the fragments, low Z materials should be used as stopping materials. This was achieved in our experiments by mixing the fissioning material with potassium chloride and pressing the mixture into a pellet.

The <sup>248</sup>Cm source used in the experiment was prepared at Argonne National Laboratory. A sample of 5 mg of curium, in the form of chloride, was first chemically purified and then precipitated as oxalate. The curium oxalate was then heated in oxygen in a furnace at 700 °C to decompose it into oxide. The curium oxide was mixed with 64 mg of KCl, was ground, and was pressed into a 7-mm diameter pellet. A pure curium oxide sample, prepared in the same way as this sample, was used for powder neutron diffraction measurement<sup>15</sup>. The measurement showed that the average particle size was  $\sim 200$  angstrom. Thus the fission fragments lost very small energy in the curium oxide and lost almost all their energies in KCl.

### 2.2. Gamma-ray Measurements

The measurements described here were made with the EUROGAM II array at Strasbourg. The device at the time of the experiment consisted of 52 large Ge detectors in anti-Compton BGO shields. Of these detectors, 28 were conventional coaxial detectors and 24 were segmented Ge detectors, called "Clover" detectors<sup>16</sup>. Each Clover detector consisted of 4 separate Ge detectors packed together in one can and it was used as a Compton polarimeter. Four LEPS detectors were also present in the array to measure X-ray spectra and low-energy  $\gamma$  rays. Approximately  $2 \times 10^9$  three- and higher-fold coincidence events were collected during the 10-day experiment. By unfolding higher-fold coincidences, a data set consisting of approximately  $2 \times 10^{10}$  triple-coincidence events were obtained. The events were sorted into  $\gamma$ - $\gamma$ - $\gamma$  and  $\gamma$ - $\gamma$ -LEPS coincidence cubes which contained  $1.8 \times 10^{10}$  and  $2.0 \times 10^9$  triple coincidence events, respectively. The energy range on the Ge axis was 3 MeV and for the LEPS, the range was 500 keV.

From these cubes,  $\gamma$ -ray spectra gated by two  $\gamma$  rays in one fragment or one  $\gamma$  ray in one fragment and the second  $\gamma$  ray in the complementary fragment were generated. On the basis of various coincidence relationships level schemes of nuclei were constructed. Two kinds of results were obtained from these studies. For many nuclei, in which few levels in the ground band were known from previous measurements at Berkeley or from  $\beta^-$  decay studies, level schemes were considerably extended and spins and parities of levels were determined.

We have also developed a new technique of analysis to determine structures of nuclei for which nothing was known previously. This was done by making a correlation of the unknown mass with the weighted average mass of the complementary fragments. This can be illustrated<sup>17</sup> by the case of <sup>103</sup>Zr and <sup>104</sup>Zr. Candidate transitions in very neutron-rich, previously unknown Zr isotopes, were obtained by gating on transitions in <sup>142</sup>Ba (Fig. 3a). Transitions which were not matched with the known transitions in <sup>142</sup>Ba and <sup>98</sup>Zr-<sup>102</sup>Zr were identified as possible transitions in <sup>103</sup>Zr and <sup>104</sup>Zr. Gating on a candidate transition in

$^{104}\text{Zr}$  generated a spectrum (Fig. 3b) which contained  $\gamma$  rays in  $^{104}\text{Zr}$  and transitions in several Ba isotopes. From the observed  $2^+ \rightarrow 0^+$  intensities of the Ba isotopes, a weighted

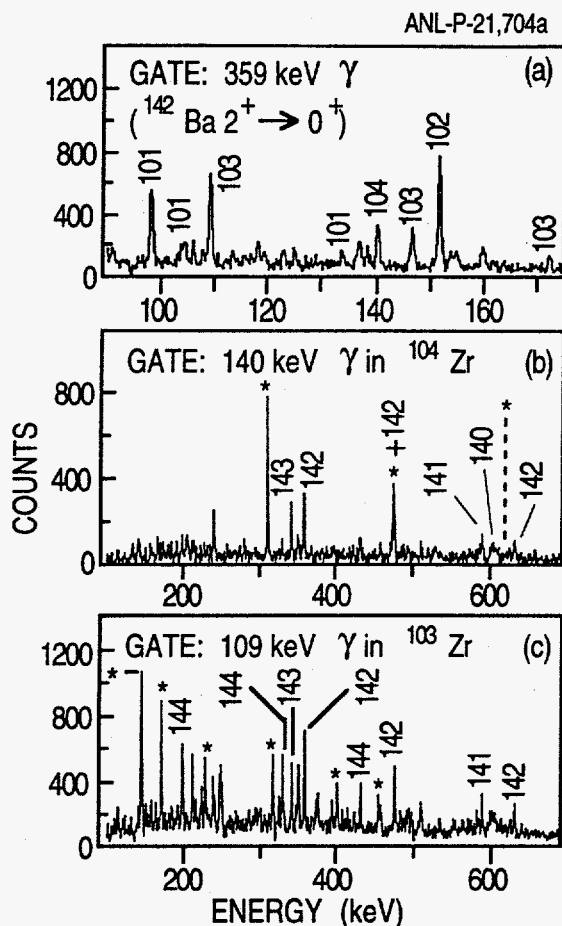


Fig. 3. Panel a shows  $\gamma$  ray spectrum gated by the  $2^+ \rightarrow 0^+$  transition in  $^{142}\text{Ba}$ . b) and c) display spectra obtained by placing gates on  $^{104}\text{Zr}$  and  $^{103}\text{Zr}$  lines, respectively. The numbers on peaks denote the mass numbers of Zr and Ba isotopes.

average Ba mass was deduced. This was done for all known and probable Zr isotopes and the weighted Ba mass was plotted (Fig. 4) against the Zr mass producing almost a straight line. The fact that the unknown mass numbers fall on this smooth curve provides evidence about the mass numbers of the Zr isotopes. The mass assignment was further confirmed by the even-even or odd-mass nature of the level scheme.

The results obtained with the EUROGAM II array are superior to the data accumulated in previous experiments. In the present run, we were able to deduce transition multiplicities of the strong transitions in many nuclei. In many cases, where the transition energy is low, the K- X-ray intensity to  $\gamma$ -ray intensity ratios gave the values of K conversion coefficient and thus the multiplicity. For other nuclei, spins and parities of levels were deduced by combining the results of triple angular correlations<sup>18</sup> and directional linear polarization measurements<sup>16</sup> of strong transitions. Another important result from these analyses is the quadrupole moments of medium spin states determined from the measured level lifetimes. The level lifetimes are determined by the Doppler-profile method, in which the observed Doppler-broadened lineshapes are fitted with a model. The details of the procedure are described in refs. 19 and 20.



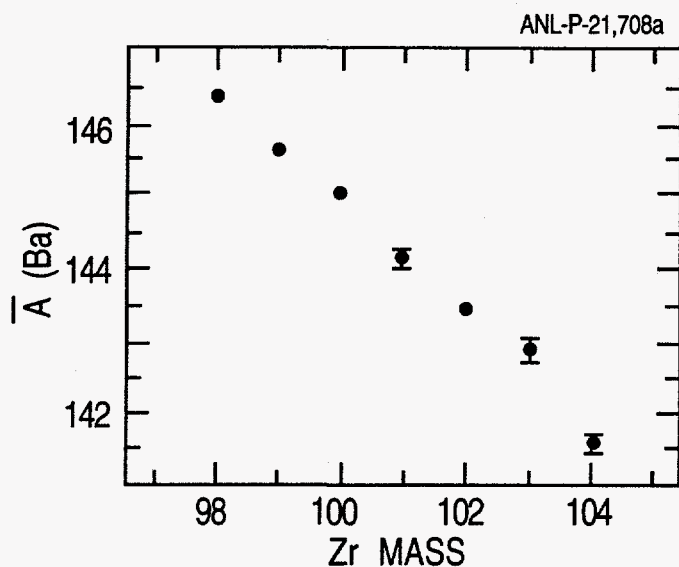


Fig. 4. A plot of the mass of the Zr fragment against the transition intensity weighted average mass of the Ba isotopes.

### 3. Discussions

Spectroscopy of prompt  $\gamma$  rays from fission fragments with large Ge arrays has provided a wealth of information on the structures and shapes of very neutron-rich nuclei. These include:

1. The elucidation of octupole deformation<sup>21-24</sup> in nuclei around mass 144.
2. The identification of single-particle states in mass 100 nuclei<sup>17</sup>.
3. The identification of two-quasiparticle states in the mass 100 region and the deduction of the pairing strength<sup>25</sup>.
4. The observation of features characteristic of large gamma deformation in Ru isotopes<sup>26</sup>.
5. The observation of two-phonon band<sup>27</sup> in <sup>106</sup>Mo and the harmonic nature of the band.
6. The quadrupole moments of medium spin states in Nd isotopes<sup>19</sup> and in the mass 100 region<sup>20</sup>. These measurements provide information on shape changes as a function of spin.
7. The elucidation of the structures<sup>28</sup> of nuclei around doubly-magic nucleus <sup>132</sup>Sn.

Below we discuss the structures of several neutron-rich nuclei in which the measurements provide evidence for the presence of octupole and gamma deformations.

### 3.1. Octupole Deformation

In the early eighties, some theoretical calculations predicted<sup>8,9</sup> possible occurrence of octupole shape<sup>29</sup> in nuclei around  $^{146}\text{Ba}$ . This was soon confirmed<sup>6</sup> by the first experiment on fission fragments with a large Ge array. The early calculations also indicated that  $^{145}\text{Ba}$  should be octupole deformed. Until recently data on odd-mass Ba were not available. In the past year, structures of odd-mass Ba isotopes were deduced<sup>21,22</sup> from the data with larger arrays. The level structure of  $^{143}\text{Ba}$  is shown in Fig. 5. Above spin 15/2, one can see a

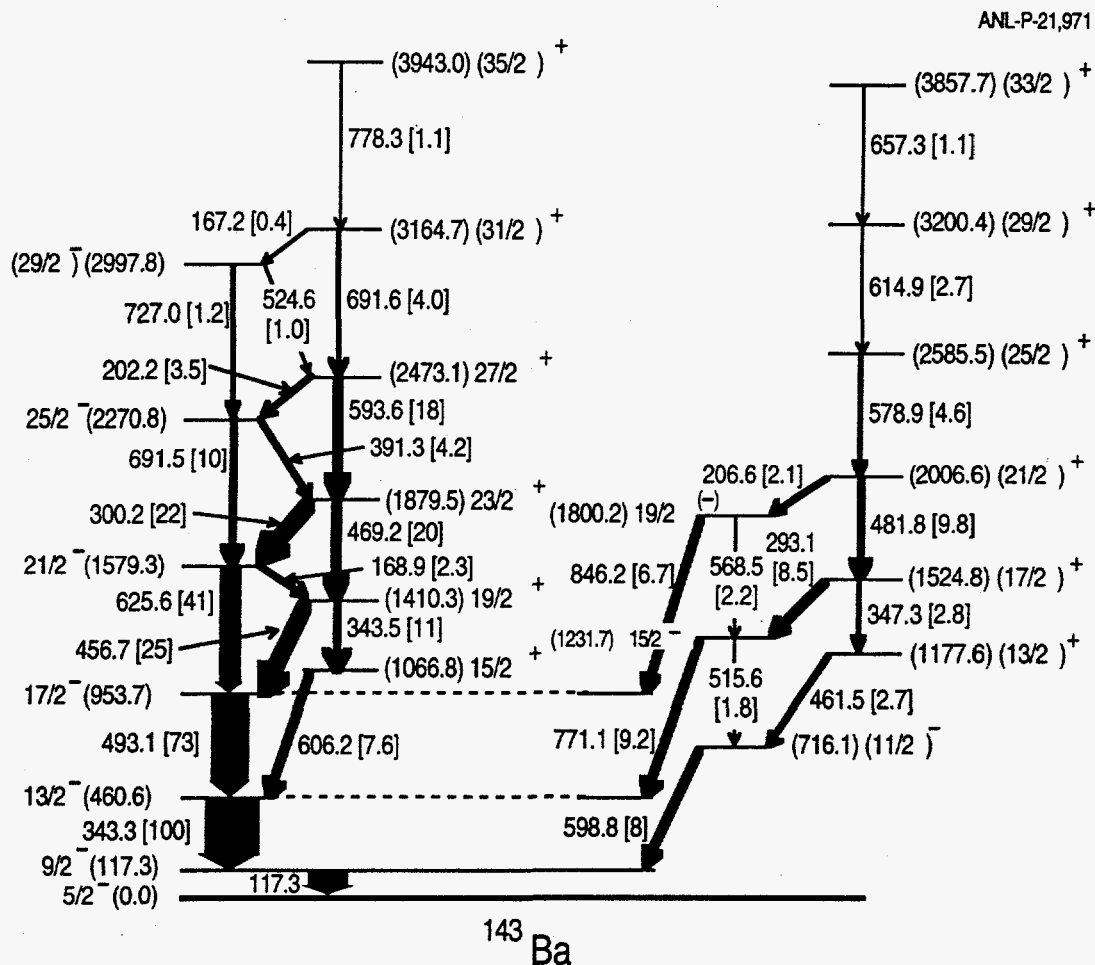


Fig. 5. Partial level scheme of  $^{143}\text{Ba}$  (Jones et al).

sequence of alternating negative and positive parity levels connected by E1 transitions. It should be pointed out that the spins and parities of levels in  $^{143}\text{Ba}$  are based on triple angular correlations and linear polarization measurements<sup>22</sup>. We see a well defined parity doublet in  $^{143}\text{Ba}$ .  $^{145}\text{Ba}$  and  $^{147}\text{Ba}$ , on the other hand, do not show level structures characteristic of octupole deformation. The dipole moments of these nuclei have been deduced from the E1 transition rates and these have been reproduced by theoretical calculations of Butler and Nazarewicz<sup>30</sup> as shown in Fig. 6. The observed dipole moments are also in agreement with calculations of Egido and Robledo<sup>31</sup> which were made by microscopic Hartree-Fock method.



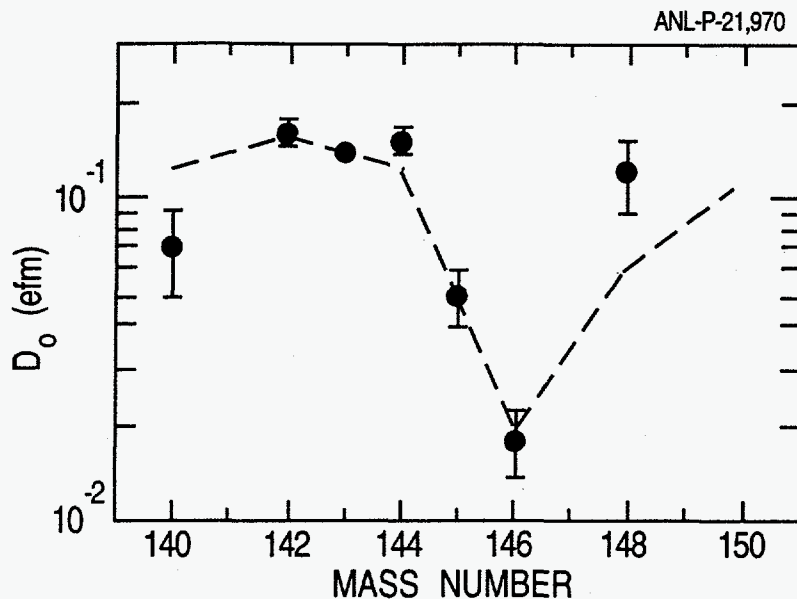


Fig. 6. Values of dipole moment  $D_0$  of Ba isotopes<sup>22,23</sup>. The dashed line represents the theoretical predictions of Butler and Nazarewicz<sup>30</sup> for the even-even Ba isotopes.

The level structures of  $^{140-144}\text{Xe}$  have also been determined from these studies<sup>24</sup>. The data on the Xe isotopes do not show any evidence for stable octupole deformation.

### 3.2. Nuclear Shapes in the Mass 100 Region

A large amount of data has been obtained from the studies of fission fragments. The measurements of fission fragment prompt  $\gamma$  rays as well as the measurements of  $\gamma$  rays following the  $\beta^-$  decay of the fragments show shape changes in the mass 100 region<sup>1,32</sup>. These shape changes are predicted in several theoretical calculations<sup>33-36</sup>. In general, the light isotopes of Sr-Zr are found to be near-spherical whereas the heavier isotopes are prolate deformed. Using our new technique, we have identified<sup>17</sup> transitions in many nuclei about which nothing was known previously. For example, we have identified transitions in  $^{104}\text{Zr}$  and deduced its quadrupole deformation to be  $\sim 0.43$ , which is one of the largest deformations known for any normal-deformed nucleus. In  $^{101}\text{Zr}$  and  $^{103}\text{Zr}$ , single-particle states were identified which shed light on the deformation in these nuclei.

Theoretical calculations<sup>36</sup> predict that some of the nuclei in the mass 100 region should have large amount of triaxiality. We have deduced<sup>26</sup> the level schemes of even-even  $^{108-112}\text{Ru}$  from the EUROGAM run. Figure 7 shows the level diagram of  $^{108}\text{Ru}$ . We have derived the ratios of the reduced electric quadrupole transition rates between low-lying levels which agree well with the predictions of Day and Mallmann<sup>37</sup>. In the calculations of Day and Mallmann, the  $B(E2)$  ratios depend on the quadrupole deformation  $\epsilon$  and the triaxiality parameter  $\gamma$ . The ratios of the measured and calculated  $B(E2)$  values are plotted in Fig. 8. The excellent agreement between the two quantities provides evidence for triaxiality in  $^{108}\text{Ru}$ . The best agreement was obtained with the ratios calculated with  $\gamma = 22.5^\circ$ . This is one of the largest measured gamma deformations for any nucleus.

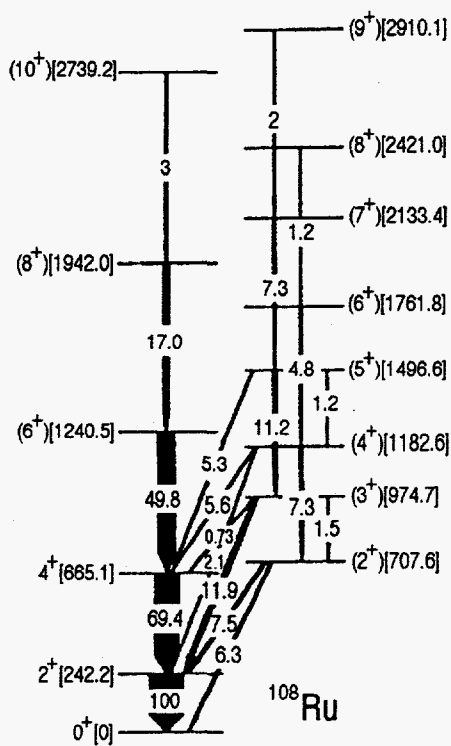


Fig. 7. Partial level scheme of  $^{108}\text{Ru}$  (Shanon et al.).

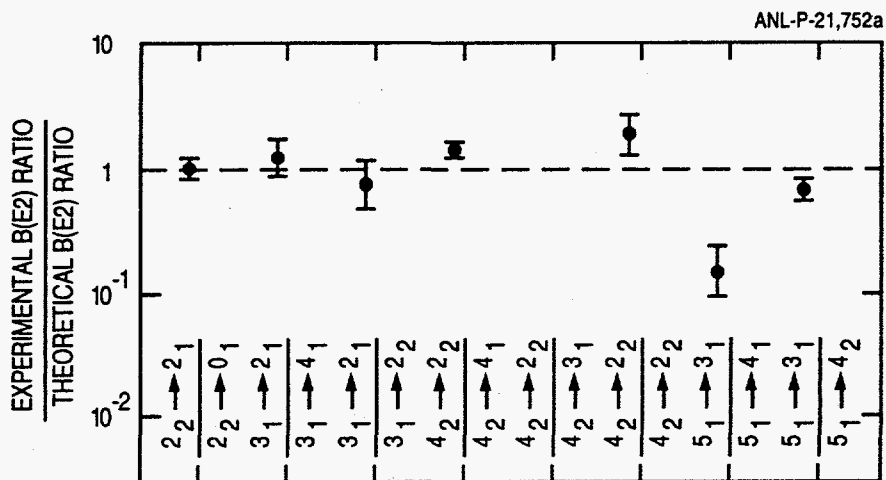


Fig. 8. Plotted are the ratios of experimental and calculated  $^{37}\text{B(E2)}$  branching ratios for transitions from various levels in  $^{108}\text{Ru}$ .

## Summary:

Prompt  $\gamma$  rays from spontaneous fission sources have been measured with large Ge arrays. These measurements provide information on nuclear shapes in many fission products. Triple and quadruple coincidences produce extremely clean  $\gamma$ -ray spectra and allow measurements of structure of nuclei with yields as low as 0.1%. The detector configurations allow measurements of directional correlations and linear polarization which are used to deduce spins and parities. In the case of the EUROGAM run, it was possible to deduce level lifetimes from the Doppler profile of line shapes. These analyses provide information on the evolution of shapes as a function of spin.

## Acknowledgments

This work was supported by the US Department of Energy under contract No. W-31-109-ENG-38 and by the Science and Engineering Research Council of the UK under grant no. GRH71161. The authors are also indebted for the use of  $^{248}\text{Cm}$  to the Office of Basic Energy Sciences, U.S. Department of Energy, through the transplutonium element production facilities at the Oak Ridge National Laboratory.

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