Spectroscopy of Neutron-rich Nuclei Populated in the Spontaneous Fission of ^{252}Cf and ^{248}Cm

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INTRODUCTION

Spontaneous fission provides a very natural production mechanism for neutron-rich nuclei. The large excesses of neutrons over protons in heavy systems near the valley of stability, coupled with the tendency for the charge-to-mass ratio to be preserved in the fission process, results in fission fragments that lie well to the neutron-rich side of stability. The excitation energy of the primary fragments produced at scission results in the evaporation of neutrons (Fig 1) to produce secondary fragments whose excitation energy is dissipated through γ -ray emission. In the last few years the study of such γ rays has produced a dramatic increase in the available information on the structure of neutron-rich nuclei. This renaissance in fission-fragment spectroscopy has been due primarily to the much increased resolving power of large arrays of germanium γ -ray detectors such as Euroball and Gammasphere. Through the improved detection efficiency and high granularity, these arrays have made it possible to obtain reasonable rates for the detection of three coincident γ rays out of the cascades of multiplicity ten that are typically produced in spontaneous fission. In addition to the determination of the energies of excited levels, developments in spectroscopic techniques have allowed for information to be deduced regarding the spins and parities [1,2] and lifetimes [3,4] of such states of extreme isospin.

In this paper we present research that has been carried out using the Euroball and Eurogam arrays to detect γ rays emitted from spontaneously fissioning ^{248}Cm and ^{252}Cf . The paper focuses on three sub-areas of current activity, namely, the measurement of yields of secondary fragment pairs, the measurement of state lifetimes at around spin 10, and recent measurements of g-factors of excited states in fission fragments.

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FIGURE 1. A schematic illustration of the process of spontaneous fission, with ^{248}Cm as the fissioning system.

I YIELD MEASUREMENTS

Lately there has been significant interest in the secondary yield measurements for ^{252}Cf fission generated by the observation [5] of larger-than-expected yields for fragment pairs corresponding to the emission of high numbers (8-10) of neutrons in the Ba/Mo split. The yield distributions for Ba's with Mo's have been interpreted in terms of a bimodal fission process, of which the low TKE (Total Kinetic Energy) mode has been associated with a highly deformed configuration of the nascent Ba fragments. We have undertaken a measurement of the yields of complimentary Ba and Mo fragments using the ^{252}Cf data set (comprising 10^{10} events of at least three coincident γ rays). The method requires the accurate determination of the intensities of the often-weak coincidences occurring between γ rays emitted from fragment pairs corresponding to different numbers of emitted neutrons. To fit the required coincidence peaks in an even-handed way, we opted to sort the data into a γ - γ - γ cube which was then searched and fit using the AMPSAF [6] software using a local projection method for the determination of background. The result of this fit was a database of photopeak centroid and intensity information which was then interrogated for the coincidences of interest. In Fig 2 the AMPSAF database was gated on the the $2 \rightarrow 0$ transition in ^{144}Ba and pairs of γ rays were used to determine the intensity of each

FIGURE 2. Yields of Mo isotopes observed in coincidence with ^{144}Ba

Mo isotope. Fig 3 shows results for which ^{104}Mo was isolated in a spectrum composed of a double gate on $2 \rightarrow 0$ and $6 \rightarrow 4$, added to a double gate on the $4 \rightarrow 2$ and $6 \rightarrow 4$ transitions. In this combined spectrum, the intensities of low-lying transitions in the Ba isotopes were used to give the secondary-fragment yields of Ba isotopes in coincidence with ^{104}Mo . Special care was required to isolate ^{104}Mo from ^{108}Mo , which has similar energies for the $2 \rightarrow 0$ and $4 \rightarrow 2$, hence our use of double gates involving the $6 \rightarrow 4$ transition. Superimposed on the measured distributions is a discretised Gaussian function with width $\sigma = 1.08$ neutrons. This curve provides a good representation of the distribution of $(\nu - \bar{\nu})$, where ν is the number of emitted neutrons, and $\bar{\nu}$ is the mean, for a variety of low energy fission processes [7]. It can be seen that the measurements of secondary-fragment yields presented here are in reasonable agreement with the trend of this distribution. The results of our preliminary analysis for these and other Ba/Mo combinations have so far failed to provide substantiating evidence of the low TKE mode.

II LIFETIME MEASUREMENTS

In this work the spontaneously fissioning isotope ^{248}Cm was used as a source of neutron-rich fission fragments whose electromagnetic decay properties were to be studied. The source consisted of about 5 mg of curium oxide (giving a fission rate of about 7×10^4 per second) embedded in a pellet of potassium chloride. The pellet was large enough (0.5mm thick and 5mm in diameter) that the fission fragments were stopped within it. The Eurogam phase 2 array of Compton-suppressed germanium detectors was used to detect the γ rays emitted in the decay of the excited fission fragments. Around 2.5 billion coincidence events with more than three γ rays were recorded on magnetic tape. From these data a $\gamma\gamma\gamma$ cube and a $\gamma\gamma\gamma\gamma$ hypercube [8] were constructed. To obtain spectra that emphasised a particular decay sequence, for the most part triple-gated 1D spectra from the hypercube were used, though in a few cases where the statistics were poor, double-gated 1D spectra from

FIGURE 3. Yields of Ba isotopes observed in coincidence with ^{104}Mo

the cube were used.

At spins of $I \approx 10$, symmetrically Doppler-broadened lineshapes are observed in the γ -ray energy spectra of many of the light fission fragments. The broad lineshapes correspond to decays from states that have lifetimes comparable to (or faster than) the stopping time (1-2 ps) of the fission fragments in the source pellet, the broadening being due to the variable Doppler-shift that is observed for the time distribution of decays from such states.

The Doppler-profile method (DPM) [3] combines a simulation of the stopping of the isotropically-directed fission fragment with a simulation of the electromagnetic decay to generate a lineshape that can be compared directly with the data and thereby extract state lifetimes. The stopping of the fragment in the KCl pellet is simulated with a Monte-Carlo computer code using electronic and nuclear stopping powers given by the computer code ZBL [9]. The initial fragment kinetic energy distribution was assumed to be Gaussian with its centroid taken from Ref. [10] (with a Coulomb correction for the particular charge split) and width taken to be the same as that measured [11] for the light fragments from ^{252}Cf spontaneous fission. The simulation produced a two-dimensional data matrix (Fractional Doppler Shift, or FDS matrix) whose contents gave the probability of observing a particular fractional Doppler shift ($\beta \cos \theta$, where β is the velocity of the fragment and θ is the angle between the fragment velocity and the γ -ray detection axis) in the emitted radiation at any given time.

In modelling the electromagnetic decay of the excited fission fragment, consideration must be given to the time distribution of the intensity feeding the states whose lifetimes are to be determined. For the highest-spin state whose decay produces an observed lineshape it is not possible to extract any information directly from the γ -ray spectrum about the feeding distribution, so the feeding of this state is simulated by a two-state feeding model in which the lifetimes of both feeding states are varied in the fit. For all other states there is some information regarding the feeding distribution contained in the lineshape of the decay into the state, although a large fraction (40-60%) of the population of the state is provided by unobserved "side-feeding"

FIGURE 4. Observed lineshapes for the $8 \rightarrow 6$ and $10 \rightarrow 8$ transitions in ^{112}Ru . The spectrum is produced from a hypercube, gated on the $2 \rightarrow 0$, $4 \rightarrow 2$ and $12 \rightarrow 10$ transitions, thereby reducing the sensitivity of the resulting Q to uncertainties in the time distribution of the unobserved side feeding.

transitions. Measurements of γ -ray multiplicities following spontaneous fission suggest that a yrast state at intermediate spin is fed predominantly by a combination of fast statistical γ rays, and the transition from the next-highest yrast state. Although slow side-feeding components have been observed they usually constitute only a few percent of the decay intensity. The time distribution of the side-feeding was simulated in the fitting procedure by a two-state model with a variable feeding time. To reduce the effect of possible contamination of the lineshapes with sharp lines from the heavy fragments the rotational band (between $I=6$ and $I=12$) was assumed to correspond to the rotation of a nucleus of constant intrinsic quadrupole moment (Q). Within this model, the mean lifetime (τ in ps) of a state of spin I , decaying within a rotational band by a stretched $E2$ γ ray of energy E_γ (in MeV) is given by

$$\frac{1}{\tau} \approx 1.217 \langle I, K, 2, 0 | I - 2, K \rangle^2 Q^2 E_\gamma^5,$$

where $\langle I, K, 2, 0 | I - 2, K \rangle$ is a Clebsch-Gordan coefficient and Q is measured in e barns (eb). Q was used to vary the state lifetimes in a manner consistent with this assumption, the best fit to the data resulting in a solution for Q . The quantum number K represents the projection of the intrinsic angular momentum on the symmetry axis. It is well defined only in the case of axially-symmetric nuclei at low rotational frequencies. In all even-nuclei examined here, K was taken as zero, whereas for the odd nuclei the bandhead value of K was used.

Fig 5 shows the quadrupole moments deduced from lifetime measurements for the light fission-fragments. The onset of deformation in the Sr and Zr isotopes is clearly seen, as is the trend to lower quadrupole moments with increasing Z . The quadrupole moments of the even Mo isotopes are lower than expected, when compared to either predicted ground-state quadrupole moments, or to those deduced from measurements of lifetimes

DPM Results

Light Fragments

FIGURE 5. A compilation of quadrupole moments deduced from our DPM lifetime measurements for rotational bands in $A \sim 100$ fission fragments

for the first 2^+ states. This difference between the quadrupole moments at low and intermediate spin has previously been interpreted in terms of a change in the triaxiality due to $h_{11/2}$ neutron alignment [4]. In the data presented here, it appears that the quadrupole moments of the odd Mo isotopes show greater stability with increasing spin, having values similar to the Zr isotopes at $I \sim 10$. All of the measurements made in the odd Mo and Ru nuclei correspond to bands of $h_{11/2}$ character and it may be that the occupation of the moderately prolate-driving $h_{11/2}$ orbitals by the odd neutron is sufficient to stabilise the axial symmetry. However, detailed calculations are required to determine whether such an effect would reproduce the observed phenomenon.

MAGNETIC MOMENT MEASUREMENTS OF EXCITED STATES

To date there have been no measurements of the g -factors of excited states in fission fragments using prompt γ rays, although there have long been experiments that measure g -factors of states populated in the beta-decay of mass-separated fission fragments. The advantage of being able to use the prompt γ rays as opposed to those emitted following β decay is two fold. Using the prompt γ rays allows access to yrast states up to intermediate spin ($8-16\hbar$) whereas post- β -decay γ rays are generally emitted from states whose spin is close to that of the parent nucleus. In addition, the use prompt γ rays may allow access to more neutron-rich species with shorter β -decay lifetimes. In this section, we describe an experiment to measure for the first time, the g -factors of excited states in neutron-rich fission fragments through the time-integral perturbed angular correlation functions between pairs of prompt γ rays.

FIGURE 6. Geometry used in the discussion of the measurement of Larmor precessions caused by the hyperfine magnetic field B

The experiment involved the use of a ^{252}Cf source of total activity $120\mu\text{Ci}$ sandwiched between two layers of Gd metal. Prior to the deposition of the Cf, the Gd metal foils (each 20mgcm^{-2} thick) were annealed in an oven at 650°C for ten minutes. The magnetisation of these foils as a function of temperature and applied field was measured using a magnetometer. The results of this measurement showed that the magnetic moment of the Gd reached 87 percent of its calculated maximum value with the temperature held at 80K. The Cf was then electro plated onto the surface of one of the Gd foils and a layer of indium metal ($200\mu\text{gcm}^{-2}$ thick) evaporated over a second Gd foil. The layer of indium acted as an aid to adhesion between the active foil and the second foil which was rolled on top to produce a closed source in which the fission fragments stop in Gd. The source was placed in a specially designed chamber at the centre of the Euroball array. In this chamber the source was clamped between a Cu strip and an Al plate, the Cu strip being attached to a cold Cu block that was maintained at around 86K by a constant flow of liquid-nitrogen. A pair of small permanent magnets applying a field of 0.2 Tesla were placed either side of the source in the direction normally reserved for the beam to Euroball. The direction of the applied field could be reversed by rotating the magnet assembly through an angle of 180° . A system of colour-coded LED's with optical-fibre links to the outside of the Euroball array was used to confirm the orientation of the magnet assembly. The experiment ran for 14 days with the field direction being changed every two hours and the data written in event-by-event mode to magnetic tape. After presorting, which involved adding together the energies of Compton events in the composite cluster and clover detectors, as well as software-gainmatching the energy signals for the whole array, 9.9×10^9 events of fold 3 or greater were recorded.

In determining the magnitude and sign of the Larmor precession for a complex array such as Euroball, it is important to combine the data from many different detector pairs. In a coordinate system Fig 6 where the applied field is along the z-axis, θ_1 and θ_2 represent the polar angles of the two detection axes respectively and ϕ is the azimuthal angle between the detection axes, we take the Larmor precession to be of an angle $\delta\phi$, about the z-axis. The effect of the rotation on W (the known angular correlation function) is then given by,

$$\frac{\partial W}{\partial \phi} \delta\phi_r = - \frac{\partial W}{\partial \cos\theta} \sin\theta_1 \sin\theta_2 \sin\phi \delta\phi_r.$$

From this expression it is clear that there are two geometric parameters that have to be recorded in any event if the data from different detector pairs are to be combined, namely, $c_{12} = \cos\theta$ and $s_{12} = \sin\theta_1 \sin\theta_2 \sin\phi$. The procedure adopted in the analysis described here was to divide Euroball into 256 bins in the space of c_{12} versus s_{12} and to label each bin with an integer index, $j(1,2)$. Since the effect of a precession on the count rate in a detector pair goes to zero as $s_{12} \rightarrow 0$, data were only used for detector combinations with $s_{12} > 0.1$

$$\rho_{ij} = \left[\frac{\uparrow N(c_{ij}, s_{ij}) \downarrow N(c_{ij}, -s_{ij})}{\downarrow N(c_{ij}, s_{ij}) \uparrow N(c_{ij}, -s_{ij})} \right]^{1/2}$$

$$X = \frac{(\rho_{ij} - 1)}{(\rho_{ij} + 1)} \frac{1}{s_{ij}} = \frac{W'(c_{ij})}{W(c_{ij})} \delta\phi$$

FIGURE 7. An illustration showing the symmetry used in producing the double ratio ρ_{ij} . An interchange of the γ rays going into detectors i and j produces an equal, but opposite, change in the coincidence count rate and corresponds to a change in sign of the parameter s .

The large number of different nuclei populated in fission means that in an experiment such as this (where the fission-fragments are not directly detected), it is necessary to use at least three specified γ -ray energies to gain adequate selectivity of the decay path. Since the angular correlation measurement concerns only two γ rays, a third γ ray is used as a so-called 'isotropic' selector. To allow for flexibility in the application of gates on various transitions, it was decided to sort the data into a 4-dimensional, high-resolution hypercube [8], whose axes corresponded to, $e_0, e_1, e_2, j(1,2)$ (e_0 is the energy of the isotropic selector, and the axis $j(1,2)$ is the angle index for the detection of the γ rays with energies e_1 and e_2). Data for both field directions were stored in the same hypercube, differing only in the allocated range on the 4th axis, thus ensuring that the applied gates were identical for the field-up and field-down data. Uncertainties caused by normalising the field-up to the field-down data, and concerns about possible changes in the detector efficiencies between runs are dealt with through the use of double ratios,

$$\rho(c_{12}, s_{12}) = \left[\frac{\uparrow N(c_{12}, s_{12}) \downarrow N(c_{12}, -s_{12})}{\downarrow N(c_{12}, s_{12}) \uparrow N(c_{12}, -s_{12})} \right]^{1/2},$$

where $\uparrow N(c_{12}, s_{12})$ represents, for example, the number of counts with field up recorded in detector combinations with c_{12} and s_{12} values identified through the index $j(1,2)$. The double ratio as defined above makes use of the symmetry that for small precessions, the fractional change in the number of counts in any detector pair is equal and opposite if the γ rays are interchanged, i.e. if $(c_{12}, s_{1,2}) \rightarrow (c_{12}, -s_{1,2})$. The double ratio can then be used to calculate the quantity,

$$X(\cos\theta) = \left[\frac{\rho(c_{12}, s_{12}) - 1}{\rho(c_{12}, s_{12}) + 1} \right] \frac{1}{s_{12}} = \frac{1}{W} \frac{dW}{d(\cos\theta)} \delta\phi_r,$$

FIGURE 8. Plot of the mean value of X against the cosine of the angle between detectors, for the first 2^+ state in ^{144}Ba . Superimposed on the data is a fit proportional to the logarithmic derivative of the measured angular correlation function between the $4 \rightarrow 2$ and $2 \rightarrow 0$ transitions.

a product of the logarithmic derivative of the angular correlation function and the precession angle. In the data analysed here, a weighted mean of $X(\cos\theta)$ is taken over all the detector combinations corresponding to a given $\cos\theta$, and the results fitted with the known logarithmic derivative of W to extract the precession angle.

Figure 8 shows a measured precession in ^{144}Ba , using the correlation between the $2^+ \rightarrow 0^+$ and $4^+ \rightarrow 2^+$ yrast transitions. The isotropic gate is placed on the $6^+ \rightarrow 4^+$. Plotted in the figure is $X(\cos\theta)$ against $\cos\theta$, from which the magnitude and sign of the precession are obtained. Several such measurements are possible for the $I^\pi = 2^+$ state, just by choosing a different isotropic selector, or by using, for example, the correlation between the $2^+ \rightarrow 0^+$ and the $6^+ \rightarrow 4^+$ γ rays. Fifteen different gates were analysed to obtain the resulting precession of 47(7) mrad. Using the known g-factor [12] and lifetime [13] for the ^{144}Ba 2^+ state, it is then possible to determine the static hyperfine field experienced by the implanted ions, using an approximate formula valid for small precessions,

$$\delta\phi_r \sim 47.8gB\tau$$

where the angle is in mrad, the field (B) in Tesla and the state lifetime (τ) is in nanoseconds. This gives the hyperfine field for implanted Ba nuclei as 2.8(4) Tesla. It should be noted that the measured field is much lower than expected, typical static hyperfine fields in Gd vary with the Z of the implanted ion, and from sample to sample, but are often above 10 Tesla. Analysis of the 2^+ states in the even Zr isotopes suggests that the field strengths may be as much as 5 times smaller than have previously been measured in in-beam experiments. The cause of the low fields is not known with any certainty, but may be due to damage of the Gd lattice caused by the fission fragments in the 8 months between the source being manufactured and the experiment being performed.

In spite of the disappointing field strengths, it has proved possible to measure precessions for several states with nanosecond lifetimes. Measurements have been made for the first $I^\pi = 2^+$ states in $^{144,146}\text{Ba}$ and $^{146,148,150}\text{Ce}$ as well as for the $9/2^-$ state at 117 keV in ^{143}Ba and the $7/2^-$ state at 114 keV in ^{145}Ba . The deduced g-factors are presented in Table 1. Also shown in figure 9 is a comparison between the results

FIGURE 9. The g-factor results from this work are compared with those compiled in Raghavan. The first 2^+ states in ^{144}Ba and ^{148}Ce are used to calibrate the field strengths.

obtained here and those compiled in reference [12]. It can be seen from this comparison that in general the results obtained here are consistent with previously known g-factors, for those cases where measurements have been made. The measurement of the 2^+ state in ^{146}Ba is consistent with the previous result and provides supporting evidence for a downward trend in the g-factors of the 2^+ states of even-even Ba isotopes. It has been suggested [14,15] that this decrease may be explained within the framework of the IBA as due to the increasing number of neutron valence bosons that occurs in the first half of a neutron shell and the effective g-factor associated with the neutrons.

The measurements of the $9/2^+$ and $7/2^+$ states in ^{143}Ba and ^{145}Ba , respectively are the first measurements for these states. In figure 9 these data are compared with the known g-factors for the ground states of these nuclei. In both cases, there is no measurable change in the g-factor between the ground states and first excited states, within the rather large experimental uncertainties. The ^{150}Ce result is the first g-factor measurement in this nucleus, but due to the large experimental uncertainty, it is difficult to say whether the trend of decreasing g-factors for the 2^+ states appears also in the Ce isotopes.

III CONCLUDING REMARKS

This paper has discussed three areas of research related to fission-fragment spectroscopy. The measurement of coincidence yields of fission fragments, particularly for the Ba/Mo split, is an area of topical interest and efforts will continue in search of supporting evidence for the low TKE fission mode. The lifetime measurements for the $A \sim 100$ nuclei presented here now provide the first comprehensive survey of the evolution of nuclear shape in this region at intermediate spin, from the well deformed Sr and Zr isotopes through to the much softer Pd nuclei. Also presented in this paper has been the first attempt to measure g-factors of excited states in fission-fragments using prompt γ rays. There is considerable scope for the refinement of the techniques presented here, both for the measurement of lifetimes and for g-factors. The dividends of such developments being access to information on the electromagnetic properties of excited states in nuclei which are of the most

Nucleus	State	Lifetime (ns)	Precession (mrad)	g-factor
^{143}Ba	$9/2^-$ (117keV)	3.8 ± 1.2	47 ± 28	0.10 ± 0.06
^{144}Ba	2^+ (199keV)	1.06 ± 0.06	47 ± 7	0.34 ± 0.05 [†]
^{145}Ba	$7/2^-$ (113keV)	0.35 ± 0.1 [†]	-16 ± 11	-0.36 ± 0.25
^{146}Ba	2^+ (181keV)	1.26 ± 0.09	32 ± 16	0.20 ± 0.10
^{146}Ce	2^+ (259keV)	0.36 ± 0.06	-27 ± 19	0.46 ± 0.34
^{148}Ce	2^+ (159keV)	1.5 ± 0.1	-89 ± 17	0.37 ± 0.06 §
^{150}Ce	4^+ (306keV)	0.26 ± 0.10 [†]	-47 ± 21	0.8 ± 0.4

TABLE 1. g-factor results. [†]g-factor from Raghavan, At. Data Nucl. Data Tables 42,189(1989) used to calibrate static hyperfine field for Ba ($B=+2.8(4)$ Tesla) §g-factor from Raghavan, At. Data Nucl. Data Tables 42,189(1989) used to calibrate static hyperfine field for Ce ($B=-3.4(8)$ Tesla) [†]Estimated lifetime as no available experimental measurements.

neutron-rich that can be produced using available techniques.

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